RESUSPENSION OF ROCKY FLATS SOIL PARTICLES CONTAINING PLUTONIUM PARTICLES - A REVIEW

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ABSTRACT

This report provides an overview of research conducted at Rocky Flats Plant (RFP) dealing with the resuspension of soil particles from plutonium (Pu) contaminated soil in the 903 pad field. The following resuspension processes were considered - saltation (wind erosion of bare soil), wind resuspension of particles from grass blades, rain splash, mechanical disturbances and grass fires. Wind resuspension from bare soil seems to be minimal, while resuspension from grass appears to be the dominant process. Rain splash is also a significant resuspension process. Over 90% of the Pu is associated with soil particles larger than 3 μm. The airborne activity is roughly proportional to the mass of particles collected. Resuspension of respirable particles from the field is very limited (i.e., the concentration is near fallout levels). Transport of the Pu extends to about 1.5 km from the pad. The release of Pu is parameterized by a resuspension factor of 5×10^{-11} m^{-1} and a resuspension rate of 2×10^{-12} sec^{-1}. The total resuspension, which is very low, is estimated at 200 μCi/yr. For a typical respirable particle concentration of 10 aCi Pu-239/m^3 near the pad field, the Pu collected is equivalent to one, 1 μm Pu particle per month, when sampling at a rate of 1.1 m^3/min.

TABLE OF CONTENTS

INTRODUCTION

The public is concerned about the health hazard of the Rocky Flats Plant (RFP) 903 area pad field soil, which contains plutonium (Pu) particles. The concern is recognized by RFP, which has been monitoring this area, since the first waste cutting oil drum leaks were discovered some 30 years ago. The oil contained <3 μm Pu particles. The drums were removed by 1969 and contaminated soil was removed and the area was stabilized with an asphalt pad. During this period and continuing today, the air at the pad field and in the Denver area is continuously monitored for airborne Pu/soil particles. (In the discussions that follow, it is understood that the Pu is attached to soil host particles.)

At no time, since the 1971 completion of the drum storage clean-up, has the Pu concentration exceeded the DOE "Derived Concentration Guide" of 20 fCi/m^3, even in the source area. As a matter of fact, the concentration of respirable Pu particles, which present the health hazard in the air that has passed over the pad field, is near background levels found in the Front Range area of Colorado. Considering all pathways of Pu intake, and even if the doses are considered additive, the population exposure is well below EPA guidelines (EP78p221)[*]. Stated in another way, the alpha radiation dose to the population from the RFP Pu is insignificant compared to the naturally occurring radon. The average activity concentration of airborne Pu at the plant boundary is 0.05 fCi/m^3 vs. an average radon concentration of 110,000 fCi/m^3 in the US (NC87p12)! The Denver area radon concentration is about twice the US average. Nevertheless, the 903 pad is scheduled for investigation and remediation in the future, even though the alpha activity in the pad field soils are much less than the radioactivity found in radium tailings in downtown Denver or those in uranium mine tailings in Grand Junction. The activity from RFP Pu in environmental waste amounts to a few curies, while mine tailings activity amounts to a few thousand curies in the middle of town (KA84p130). In the city of

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[*] The last part of the literature citation, following the "p", indicates the page number at which the information will be found.

Grand Junction the radon activity is about 10 times higher than normal due to the uranium mine tailings (KA84p132).

A synopsis is provided of RFP research on the resuspension of Pu particles from the pad field. This includes delineation and activity of the source area, consideration of all feasible processes of resuspension and the subsequent transport of the airborne particles, including their size and activity. This work has been time consuming and has proved inconclusive at times, because of the low Pu activity involved.

SOURCE AREA AND SOURCE ACTIVITY

The area where the drums had historically been stored, now known as the 903 pad field (see Fig. 1), was covered with gravel and asphalt to immobilize the Pu contaminated soil particles. However, during site preparation for the asphalting, occasional high winds swept across the uncovered area. Some dust was generated and it generally settled a short distance to the east toward the security fence. This adjacent area was covered with off-site top soil and vegetation was re-established to control resuspension of the Pu particles. The fugitive Pu particles were mixed into approximately 20 cm of new top soil. This makes some Pu available for resuspension. Therefore, the area has restricted access and the air is continuously monitored in the vicinity.

It should be noted that both mine tailings from radium extraction, as found in Denver, and mine tailings used in Grand Junction for home construction, pose greater health hazards. These tailings release radon gas, which is difficult to immobilize, and the radon decay products become attached to respirable dust particles. The RFP Pu particles are relatively immobile and require mechanical force to become airborne, which by the nature of the process results mostly in particles too large to be inhaled (HA80ap216). That is, once the small (<3 μm) Pu particles are immobilized by attachment to soil particle aggregates, it is very difficult to separate the particles again, because one has to overcome the interatomic surface forces.

Fig. 1 also shows the distribution of Pu in soil at and near the plant determined by the AEC Health and Safety Laboratory (HASL) (RF3115p14). The amount of Pu that leaked from
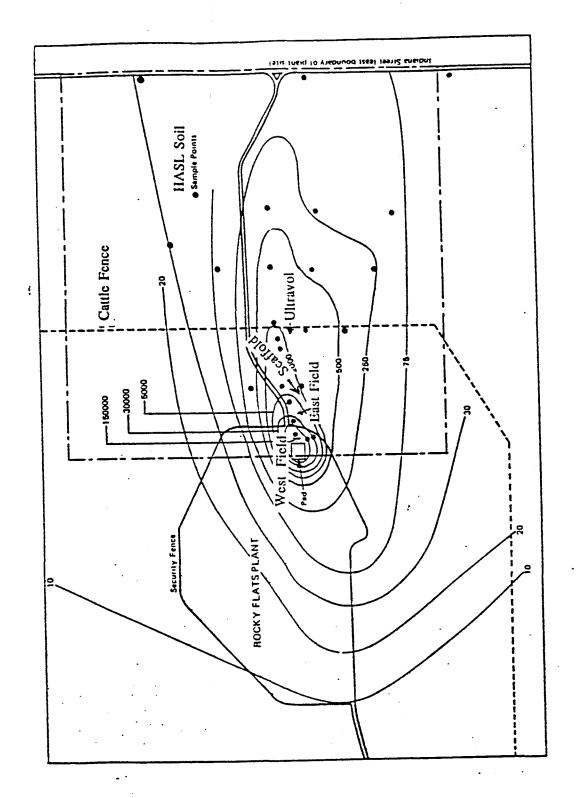
FIGURE 1. Plutonium-239 Deposition Contours in Millicuries Per Square Kilometer, According to HASL

the drums was estimated as 6.1 Ci or 86 g (SE71p6). Nearly four Ci are now under the asphalt pad. A detailed ground gamma survey (RF3689p18) for the Am-241 associated with the Pu indicate that approximately 1.2 Ci of Pu exists in the pad field area west of the perimeter fence. This survey was made after the 1978 "lip area" soil removal of an estimated 0.5 Ci Pu-239. Another 0.67 Ci exists on the other side of the fence in a small, localized area above the gunnery range. Fig. 2 gives the details of the survey. These regions of high Pu concentrations are considered the source area for chronic release of Pu from RFP and this soil is likely for future removal once an acceptable regulatory framework has been worked out.

Krey (KR76p214) has estimated, based on limited samples, that in addition to the Pu in the pad area there is another 3.4 Ci spread out over a wide region. In that case, based on a material balance, more than 9 Ci Pu-239 should have leaked from the drums, instead of the 6.1 Ci mentioned above. Alternatively, it has been suggested that some of this Pu may be traced to the 1957 and 1969 fires. This region, with near fallout Pu levels, extends east and southeast from the security fence toward the cities of Thornton and Denver. Once past Indiana Street, the eastern boundary of the plant, only a few % of the Pu is of RFP origin (i.e., the rest is from fallout). The RFP contribution in the region immediately east of Indiana Street is approximately 0.1 Ci.

An isotopic ratio (Pu240/Pu239) analysis was used by Krey to distinguish the RFP contribution from atmospheric fallout. Krey presumed that the Pu in this outlying region was due to smoke deposition from the two "Pu fires" at the plant. The fires principally involved contaminated non-Pu components, such as plastic, rubber, wastes and solvents. It should be kept in mind that the lee of the Rocky Mountains is a region of high fallout, which could include unaltered RFP Pu from the Nevada Test Site safety shots (HA80bp66). These shots release Pu particles due to the explosion of the chemical explosives in the weapon without fission taking place.
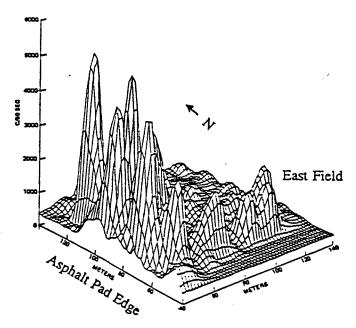
East Field

FIGURE 2A. Gamma Survey of West Side of Pad Field
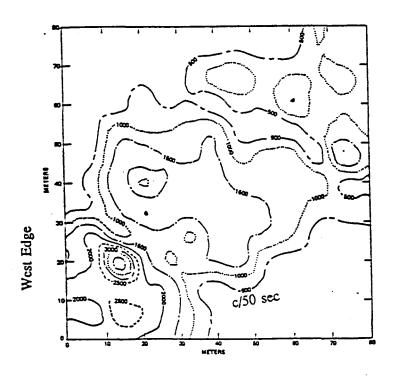

c/50 sec

West Edge

FIGURE 2B. Gamma Survey of East Side of Pad Field

Of interest are also the periodic gamma surveys of the plant and surrounding land by helicopter (BO82). Fig. 3 presents the 1981 survey contours for Am-241. The 1989 aerial survey data has not yet been published, but some of the initial results are of interest.

The sensitivity of the instrumentation was improved, making it possible to detect Am-241 beyond the cattle fence (i.e., to the 250 mCi/km^2 contour shown in Fig. 1). Also, the survey was extended beyond the confines of the buffer zone to obtain a broader picture of background activity. As far as total activity is concerned, which includes natural radioactivity sources, the highest level was found in the Jefferson County airport area and the hottest spot was an old mine shaft near Leyden, not RFP. To detect the above background activity at RFP, the survey has to be specific for Am-241, because the natural g radioactivity is too high for any RFP contribution to be seen.

The airborne surveys show that Pu activity has not migrated significantly beyond the original source area after the 1969 closure/stabilization of the 903 drum storage area.

PHYSICS OF SOIL RESUSPENSION

The potential for chronic release of airborne Pu particles from RFP is limited mostly to soil resuspension from the pad area, except the very low level Pu release from the plant's filtered production building air effluents from the plant. We know from air monitoring that Pu particles do become airborne, but the physics of the process needs to be understood for dispersion modeling and remedial action purposes. Soil particles are traditionally thought to be resuspended by wind forces acting on bare soil surfaces, but such studies have been limited to plowed fields and desert areas. The pad field has only small bare soil areas between the clumps of bunch grass, but the original premise was that the Pu particles originate from the bare soil between the clumps of grass.

Saltation

Initial resuspension studies were directed at the classical resuspension process of saltation (HA80ap213). That is, the wind propels millimeter sized particles, that protrude beyond the millimeter or so thick boundary layer (the immobile or stagnant air layer next to the surface), in a

AMERICIUM CONCENTRATION (nCi/m²)
ASSUMED DEPTH IN SOIL

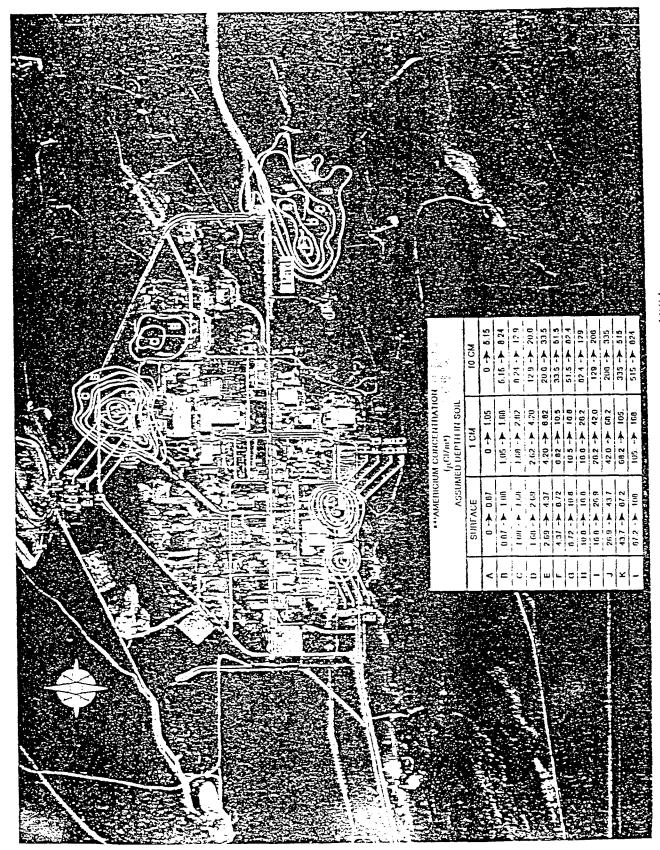|	SURFACE	1 CM	10 CM
A	0 → 0.07	0 → 1.05	0 → 6.15
B	0.07 → 1.00	1.05 → 1.60	6.16 → 8.24
C	1.00 → 1.60	1.60 → 2.62	8.24 → 12.9
D	1.60 → 2.69	2.62 → 4.20	12.9 → 20.0
E	2.69 → 4.37	4.20 → 6.82	20.0 → 33.5
F	4.37 → 6.72	6.82 → 10.5	33.5 → 51.5
G	6.72 → 10.8	10.5 → 16.8	51.5 → 82.4
H	10.8 → 16.0	16.8 → 26.2	82.4 → 129
I	16.0 → 26.9	26.2 → 42.0	129 → 200
J	26.9 → 43.7	42.0 → 68.2	200 → 335
K	43.7 → 67.2	68.2 → 105.	335 → 515
L	67.2 → 100	105. → 160	515 → 824

FIGURE 3. Aerial Gamma Survey - 1981

series of small hops. On impact these large particles knock loose smaller particles, similar to sand blasting. The smaller particles then become entrained into the main air streams by turbulence to heights over 6 m in a distance of less than 30 m (HA80ap222).

It remained for RFP to demonstrate if saltation could occur from small bare areas, because this has received little attention from soil scientists. Direct visual observations initially indicated, that even during wind storms reaching over 100 mph, no visible quantities of dust were released (i.e., puffs of dust from bare areas). Operation of a modified Bagnold Catcher for several week long runs, including windstorms, provided no weighable dust fractions. The Bagnold Catcher (GI74) is the classical device for measuring wind erosion of soil. Nevertheless, more sensitive techniques were developed to verify if saltation processes on a small scale may occur.

One technique developed by RFP was the application of a ribbon like laser beam, grazing the soil surface, to detect impacting large particles and bursts of numerous small particles (RF3197p8). This was done at night using time exposures photography. No particles were observed until winds exceeded 35 mph, but even then resuspension was tenuous and no clear evidence for saltation was found by this approach (RF3325p3).

The second methodology, shown in Fig.4, involved placing an acoustic particle detector underground, facing a 2.5 cm opening in a bare soil surface area (RF3115p11). There was no net airflow into the opening, but the objective was to detect particles over 50 μm aerodynamic equivalent diameter (AED) that were resuspended by the wind and then fell back to the ground. That is, as the particles fell back some dropped into the intake of the acoustic particle counter. This system could operate unattended for several days. Again, no convincing case could be made for saltation, even in high winds (RF3115p13,RF3197p7). Evidently, the soil is too crusty for wind erosion (HA80ap224), except for a few small areas (RF3115p11). Also, the small, bare areas are too deep in the grass canopy to experience the full force of the wind. Only deliberate disturbance of the ground (i.e., with a stick) was observed to release short bursts of particles.

FIGURE 4. Acoustic Particle Counter for Saltating Particles

*Filter is used to collect particles for microscopic exam.

The above results may seem to contradict a study by Sehmel, who developed a Pu resuspension model for the pad field based on the saltation process (SE72). However, this model was based on data collected by RFP from July 1970 through January 1971. In March 1970 the pad field was disturbed by a major ditch construction project near the west side of the fence. It took nine months for the effect of this operation to disappear (i.e., for the surface Pu to weather in again and for introduced grasses to grow).

Next, we carried out resuspension studies under controlled conditions using the small wind tunnel shown in Fig. 5 (RF3197p5). The idea was to look at resuspension from bare spots, as well as from grassy areas. Testing of bare spots showed very little release until extreme velocities (i.e., equivalent to 150 mph at 10 m) were applied or the soil had previously been disturbed. But even the latter, "fresh" surface, was quickly exhausted of particles (RF3689p36). However, this windtunnel proved to be a useful approach to soil surface sampling for Pu particles and was extensively used for this purpose (RF3689p23).

The windtunnel was also operated on patches of grass, although it was not designed for this purpose. That is, the wind tunnel test section height was less than the height of the grass. For proper testing of grass covered areas a considerably larger wind tunnel is required, such as the device of Gillette (GI78). Nevertheless, even at low velocities (i.e., wind speeds equivalent to 20 mph at 10 m), small but significant amounts of Pu were resuspended. This was considered important, since most of the ground is grass covered. Much of the resuspended material was over 10 μm AED and was organic (i.e. grass litter), as shown by ashing the samples. The organic content was about 40% on the average (RF4036p23).

Resuspension from Grass Blades

As a result of the above wind tunnel studies, attention was now focused on the details of resuspension from grass covered areas. We wanted to know the origin of the Pu resuspended from grassy areas. Does the Pu reside on the grass blades and some becomes resuspended or does Pu originate from the grass litter on the soil surface, i.e., the grass decays and then Pu is
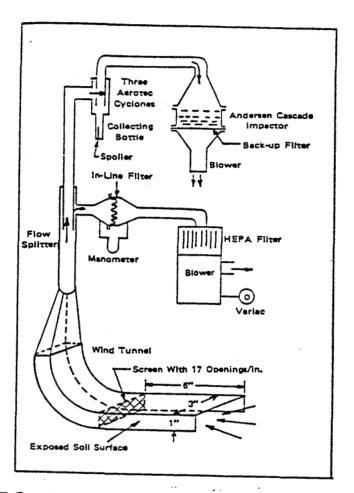
FIGURE 5. Portable Wind Tunnel for Resuspendible Dust

resuspended as part of decomposed grass particulates. Obviously, both processses could be taking place.

First, we verified quantitatively that Pu does reside on grass and grass litter. This was done by clippping the grass at successive lower levels to show the Pu distribution versus height (RF3914p6). The Pu concentration in the litter was also measured. Activity was distributed fairly uniformly vs. height for a total of 1.1×10^4 pCi from a square meter of grass grown on soil with an activity of 2200 pCi Pu-239/gm. The litter held 510 pCi Pu-239/gm. Therefore, grass blades must be considered a major source of Pu particles for resuspension and not just the litter at the base of the grass. It should be noted that the litter is not readily accessible for resuspension because it shielded from wind by the grass.

The question of how the Pu becomes attached to the grass is of interest. Plant uptake of Pu is not a factor (AR82p33). This leaves wind driven soil particles from bare soil areas and rain splash as the source of Pu. The latter process is well documented for transferring Pu to vegetation to heights up to 30 cm (DR84p183). We did not attempt to establish the importance of each of these processes. Finally, the growth process of the grass was considered as a means of transferring Pu to the blades. But the grass blades start from the stem of the plant after it rises out of the ground.

Information was now required on the capacity of grass blades to hold soil particles and on potential Pu resuspension mechanisms from the grass. Therefore, we studied the surface of grass blades with scanning electron microscopy. Interestingly, most grass blades have their surface covered with fine hairs, that act like a filter matrix that intercepts considerable amounts of dust (RF4036p15). Gutfinger (GU85p3) reports that fibrous elements extending from a surface into the viscous boundary layer enhance deposition by a factor of 10 to 1000. Conversely, dust particles should be released when the grass hairs decay and fall off and by flexing of the blades due to wind. Such behavior was verified with wind tunnel tests described below.

In a small laboratory wind tunnel (RF4036p23) samples of grass were placed in the test section and exposed to equivalent 10 m wind speeds of 5 to 20 mph. From a 5 cm long blade

about 200 particles were released in the 0.2 to 12 μm range as verified with an optical particle counter and membrane filter samples. Most pertinent were tests where the blades were mechanically flexed. Here, the release of particles >10 μm was dominant, with a median diameter of 20 μm and a maximum of 40 μm.

To verify this conclusion, a simple test with the soil resuspension wind tunnel was made in the summer with the ground soaked with water but the grass dry (RF3914p8). The object was to show how much Pu is resuspended from the grass blades alone. The blades are much more exposed to the wind than the ground. The resuspension rate was about 1/60th of that for a similar dry area at a windspeed equivalent to 80 mph at 10m. At 20 mph it was 1/40th less than at 80 mph. But these resuspension rates could still account for most of the activity observed by the air samplers, since 95% of the field is covered with grass. This data has to be interpreted with some caution, because, as pointed out above, the wind tunnel was not of an optimal design for studying resuspension from grass.

This test confirmed that release of radioactivity from grass blades alone is very important, if not dominant. Additional radioactivity exists on the dead grass litter on the ground between the standing grass, but this material is not readily available for resuspension because it is protected from the wind by the stand of grass.

Rain Splash

We still needed an explanation for Pu resuspension when the soil was completely saturated during long period of rainfall as encountered in the spring (RF3914p9). For this limited test series the Pu concentration during rainfall did not differ significantly from that during dry periods. Rain splash was therefore studied as a means of releasing Pu particles into the air. First, a laboratory wind tunnel was set-up to simulate raindrops splashing on soil under controlled conditions. Provision was made to count resuspended soil particles (if any became airborne) with respect to concentration and size utilizing an optical particle counter. This experiment showed that soil particles do become aerosolized by rain splash, if a thin water layer exits on the soil surface (RF4036p30).

Evidently, these airborne soil particles are the residual particles that remain upon the evaporation of the hundreds of small satellite droplets, that form along with big splash drops (GR73p57). The satellite droplets apparently are small enough to be carried by airflow.

The resuspension process was also confirmed in the field. A small plastic tent was built over a patch of Pu contaminated bare soil. The tent was necessary to prevent airborne Pu particles from drifting into the test area from the surroundings. Nuclear track foils were placed on the resuspended residue particles collected from the splashes to verify the presence of Pu particles. The tests showed that soil particles containing Pu did become airborne due to drop impact (RF4036p30). A thousand 5 mm rain drops resuspended 5 pCi into the air from soil with an activity concentration of 2500 pCi/g. About 500 million raindrops may fall on an area of one square meter annually. To complete this analysis, the washout (GR73p121) of resuspended soil particles by rain drops should be accounted for in a real situation. The washout effect was not present in the above single drop experiment.

Grass Fires

Another potential source of resuspended particles is the ash from grass fires. So far no fires have taken place in the pad field, but we made tests to simulate such an occurrence. Fire was set in a windtunnel to the grass collected from a one square meter area of soil, where the grass had an activity of 8.1 pCi Pu-239/g. The smoke had a total activity of 34 pCi Pu-239 (LA86p91). However, placed in perspective, the resuspension of Pu in the smoke was of such magnitude, that if the whole pad field (roughly 0.02 km^2) had burned, a person would have to inhale all the smoke to intake the equivalent of an allowable life-time body burden (about 40 nCi Pu).

Mechanical Disturbances

Mechanical disturbances were also studied as a potentially important resuspension mechanism. This includes such activities as mowing, well drilling, digging and soil removal. Mowing was considered a major interest, since it takes place every year and covers the whole area. Also, it involves disturbance of the grass and soil. While research sampling took place in

the pad field east of the fence, the grass was cut during dry conditions, which should maximize resuspension. At times the tractor would be right next to an air sampler. A statistically significant increase by a factor of 5 in the total Pu concentration was discerned during such a period in June 1981 (RF3464p4). But no change was found during a similar period in 1983 at the same location (LA86p90). Again, high variability in Pu concentrations makes it difficult to establish trends, unless many samples are taken. In June-July 1987 wells were drilled in the pad field and the nearby surveillance air samplers showed an increase by a factor of about 3 in Pu activity, but such increases are often seen during the dry summer months.

Resuspension of dust, containing Pu, from the two unpaved roads intersecting the pad field was considered. It is evident from the color and quantity of the dust collected by the samplers along the road, that much of the dust collected is resuspended from the road. A 1973 study (MI73) showed that road dust activity averaged 68 pCi/gm and remedial action was initiated (i.e., oiling, grading, etc). In 1980 another road dust evaluation was carried out. As a first step the Pu activity of the road surface soil was determined (RF3689p18). The Pu surface soil activity was surprisingly low (i.e., 4.6 pCi Pu-239/g) versus adjacent soils of 790 pCi Pu-239/g monitored a foot from the road. However, one has to consider that since 1973 the road was often graded and ballast added. It had been expected, that the adjacent soil would provide a source of Pu for easy resuspension by traffic.

A truck was again driven along the road to complete the comparison with the 1973 test and to sample a larger area than that covered by a few soil samples. The dust generated right behind a rear wheel was sampled with a hivol. The road dust was very low in activity (i.e., 6.0 pCi Pu-239/g versus 1000 and 2000 pCi Pu-239/g for two adjacent soil areas). It was concluded that the roads are no longer a significant problem. Of considerable interest was the incidental, new information, that no measurable amounts of respirable (<3 μm AED) particles are generated (RF3287p7). This was also observed during a recent dam construction project at RFP. Heavy earth moving machinery created no additional respirable dust (RF3115p6).

Such observations indicate that considerable application of force is necessary to create <3 μm AED particles. It has been noticed, that the <3 μm AED dust is always black (i.e., combustion particles generated by vaporization-condensation by vehicles, furnaces and other industrial activity). This is in line with our wind tunnel results, that soil resuspended from the pad field is very low in fines (RF3689p35) and many of the fine particles at RFP are from Denver pollution (RF3990p31).

DISPERSION OF Pu PARTICLES FROM RFP

Combining the above processes into a coherent, predictive Pu transport model is obviously beset with problems, especially with the resuspension from the 903 pad field being relatively low and of varying nature (wind, rain splash and release from grass). Data from previous studies can be used to derive conventional resuspension parameters which are commonly used to characterize emission of soil particles containing contaminants (HA80ap210). This will be done, but first additional experiments will be reported that directly measured the emission and transport, including vertical distribution of Pu particles, in the air that passes over the pad field.

Measurements of Pad Field Pu Flux to Environment

Two distinct steps were involved in this study. First, an attempt was made to measure the Pu particle flux from the pad field right at the source (i.e., a number of research air samplers were deployed at selected points in the pad field to determine the total resuspension of Pu). Second, a vertical array of samplers was installed, at some distance from the pad field, to measure the Pu particle concentration in the air that passed over the pad field.

a. Determination of Pu Flux from Source Area

For the first step, four research hivols were installed toward the perimeter areas of the pad field and an ultravol was installed near the center of the field to determine how much Pu is being released to the environment. The ultravol, operating at 260 scfm, was changed weekly or more often to provide high resolution Pu concentration data (RF3197p6). This was used for special events such as wind storms, periods of snow or rain only and fallout from nuclear weapons tests

(RF3464p5). The research hivol samplers provided <3 μm AED, 3-10 μm AED and >10μm AED fractions, corresponding to respirable, inhalable, and coarse particle sizes, respectively. The 10 μm AED fraction was collected by a cyclone pre-separator, that turned with the wind so that the intake always pointed into the wind. The efficiency of this cyclone was evaluated and one observation was that the intake efficiency was not sensitive to wind speed (i.e., the particle concentration and size distribution was unaffected by wind speed) (RF3464p34).

The results of approximately two years of sampling (RF3464p4, RF3650p1) indicated that each sampler was strongly influenced by resuspension from the immediate area, probably within <10 m. This conclusion is supported by the fact that it was not possible to find a correlation versus time for the Pu concentrations between the five samplers. This result implies that most of the Pu activity is carried by particles well over 10 μm AED, since they would settle out due to gravity at near-in distances. Unfortunately, since our primary interest was in health effects, our samplers were not set-up to routinely further fractionate the >10 μm AED particles. This was done occasionally by wet sieving the >10 μm AED particles into 44 and 74 μm sieve cuts. Freon was used for the suspension medium to minimize de-agglomeration (HA80ap219). This sieving showed that the Pu distribution is roughly proportional to dust particle mass (RF4036p22). More accurately, the specific activity of the Pu in the samples was three times higher for the inhalable and coarse fraction than for the respirable fraction. But the specific activity for the respirable fraction is small and not as accurately determined.

With the above Pu concentration data only valid for the immediate area around a sampler, it was not possible to estimate an overall Pu flux from the pad field.* The dust loadings from all the samplers were well correlated in time, but this had no relevance to Pu transport, because most of the dust originates from locations beyond the pad field. Above 30 μm AED the particles were presumed to be of local origin and were mostly vegetative litter from qualitative examination.

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* The same was true for the 3 surveillance samplers spaced across the pad field along the fence, i.e., the concentration data between the three samplers did not correlate in time (RFP3914p4). The data examined was for a 9 year period.

However, in the spring large amounts of pine pollen were blown from the foothills over the plant and were collected by the air samplers. Pollen is designed by nature to float long distance due to small airsacs. Scanning electron microscope photos showed the presence of pollen (RF3990p28), which causes a yellow-green discoloration of the coarse particle dust fractions.

Correlating the Pu data with wind speed and direction proved equally futile. Even the operation of some of the samplers in a wind directionally actuated mode provided no finer resolution of Pu transport patterns.

The tests did show that the concentration of the respirable Pu fraction, 20 aCi Pu-239/m^3, is nearly at fallout level of 18 aCi Pu-239/m^3 for June 1980 to June 1981 (RF3650p6). The >3 μm AED particle activity was 710 aCi Pu-239/m3. There is little emission of <3 μm AED particles from the pad field (RF3464p4). The >3 μm AED fraction carried 98% of the activity for the above period.

b. Dispersion of Pu by Air Passing Over the Pad Field

The above studies were followed by a sampling program using a tower erected 100 m from the eastern edge of the Pu contaminated pad field (RF3914p16). The objective was to follow the transport of Pu from the pad field. The tower had hivol samplers at 1, 3 and 10 m to sample the air that had passed over the pad field for Pu particles. The three hivol samplers had size selective inlets (SSI) with a 15 μm AED cut followed by 3-15 μm AED cut and a <3 μm AED cut. The SSI was evaluated for wind speed (1-10 mph) response by Wedding (WE82) and McFarland (MC84) in windtunnels and by RFP during high winds (50-100 mph) in the field (RF3914p4). Performance was satisfactory at low speeds, but at the high wind speeds some particles were apparently blown through the inlet without being sampled. The particle flux data from this tower represents the sum of all resuspension processes active in the source area during each two month sample period. This data provides basic information to estimate possible population exposure and translocation of the Pu particles from the source area.

This research program extended from November 1982 thru August 1985, collecting bi-monthly samples. Such a long collection period was necessary to collect sufficient Pu in each

size fraction for detection. The dust concentration data showed well defined trends with sampling height (RF4036p21). However, the respirable dust particle concentration (8.0 mg/m3) did not change with height, as expected for particles that sediment slowly. The concentration for the inhalable and coarse dust particles, ranging from 10 and 25 mg/m3 respectively, dropped off with height due to sedimentation.

The Pu concentrations (RF4036p21) for the <3µm or respirable particles was 8.8 aCi Pu-239/m^3 and for inhalable (3-15 µm) particles was 25 aCi Pu-239/m^3. These concentrations did not correlate with height, being only 3 and 10 times greater than background concentration respectively. One must keep in mind that the Pu mass for these samples represents about 1/10th part per billion. Statistically the data have to be erratic at such extremely low concentrations. For a Pu concentration of 10 aCi/m^3 (typical of the respirable Pu particle concentration at the tower), it would require the collection of just one, 1 µm Pu particle per month by a hivol operating at 1.1 m^3/min.

The concentration of 67 aCi Pu-239/m^3 in the coarse (>15 µm) particle fraction was almost a magnitude higher than that in the respirable fraction. The coarse fraction exhibits a significant decrease in activity with height by factor of 3 from 1 to 10 m.

No correlation was found with wind speed or direction for the Pu or dust concentration. This can be expected for the poor time resolution given by two month sample periods necessary to collect enough Pu for analysis.

It is obviously of interest to see how the Pu concentration changes beyond the scaffold. So the ultravol sampler was operated in June 1981 at the cattle fence, 0.5 km due east of the scaffold. The Pu concentration in the inhalable and coarse range dropped off by a factor of 20, approaching background levels (UN81). Therefore, it did not seem worth while to continue this operation at such low levels. Evidently, most of the large Pu/soil particles, that carry the bulk of the Pu activity, had settled out before reaching the cattle fence in the RFP buffer zone, long before reaching populated areas. This observation is directly supported by work of J. Hayden (HA75), who measured the size of individual Pu particles found on the soil surface from the pad

field to Indiana Street. Beyond the cattle fence he considered the stack effluent to be the primary source of the Pu particles.

c. Comparison of Our Data with Precious Resuspension Studies

It is of interest to compare the above results to Sehmel's July 1973 Pu resuspension experiments at RFP (HA80cp241). Although Sehmel's was a more elaborate Pu flux study than the study cited above, it only lasted for one month. Sehmel used three sampling scaffolds, one at the fence near the 903 pad, one at the same location as our RFP scaffold and one near the cattle fence.

It is difficult to directly compare Sehmel's data to ours, because our work covered 34 months to determine statistically significant trends in the Pu concentration at three levels at one location. Sehmel's study probed the Pu particle plume at three locations with nearly 40 samplers set for specific wind speed ranges as well as continuous operation. However, to reiterate, Sehmel's research work was only of one month duration.

There is also a problem comparing the particle fraction data. To achieve well defined particle size fractions, RFP took considerable precautions to coat the collection surfaces with adhesive. The object was to stop particle from bouncing through the cascade impactor stages onto the back-up filter (RF2866p14, RF3115p4). Sehmel did not use adhesive on his collection surfaces. He showed that 60-99% of the Pu was in the respirable range and supposedly of RFP origin (HA80cp262). We found that respirable Pu was mostly of fallout origin and it only represented 2 and 9% of the total Pu activity, based on our measurements at the tower and pad field respectively. Therefore, the Pu size trends are not comparable.

The drop off in Pu concentration with distance from the pad field fence to the second scaffold varied among samplers by a factor of 10 to 1000 in Sehmel's tests (HA80cp251). We do not have comparable simultaneous data, because we only had access to five hivol samplers. But taking data over the period 1980 to 1985, as our experiments moved east, the average Pu activity at 1 m changed from 1900 to 480, to 130 and finally to 50 aCi/m^3 moving respectively from the

pad field to the east pad field, to the scaffold and to the cattle fence. This trend represents a 40 fold reduction over 1.5 km.

Sehmel did not report a definitive relationship between wind speed and Pu activity (HA80cp244). This is similar to our research experience in this area.

d. Isotopic Ratio Determination as an Indication of Long Distance Dispersion of RFP Pu.

As a final test of whether any RFP Pu particles reach the general population, the isotopic ratio of Pu240/Pu239 was determined for a series of airborne particulate samples (RF4036p22). RFP Pu production metal has a Pu-240/239 ratio of 0.051, while fallout has a ratio of 0.163. Airborne dust samples collected at the tower showed a ratio of 0.063 and nearby soil had a ratio of 0.054. This small difference was significant, indicating that the airborne dust carried some fallout Pu as to be expected. We now need to obtain the isotopic ratios for air samples taken in various parts of the Denver region to identify the RFP contribution, if any, from pad field resuspension or stack emissions.

For the latter program we also need to take soil samples at the air sampler sites, since most background Pu (fallout Pu) now originates from resuspension of nearby soil particles (RF4036p29). Stratospheric influx of Pu is very low at present. Therefore, the isotopic ratio of the soil should be known at the air sample sites, to adjust for any RFP Pu already in the soil, in addition to the fallout Pu from past nuclear weapons tests. Nearly 20 years ago isotopic ratios were determined by Krey (KR76p213) for a limited number of soil samples in the greater Denver region to identify RFP Pu. This would also be a chance to determine if any changes occurred in these ratios. Krey's data shows that 1-2% of RFP Pu in the environment extends beyond Indiana Street.

e. Resuspension Factors for Pu Release from Pad Field

The resuspension factor (R_f) estimates the airborne contaminant concentration directly above a contaminated area and provides a means to estimate exposure or dose. R_f equals the airborne Pu concentration measured directly above a given area divided by the soil surface Pu concentration at that location. We have the necessary Pu data to calculate resuspension factors

for the 903 pad field. The soil surface Pu concentration can be is derived from the soil density and soil activity per unit mass.

Another resuspension parameter is the resuspension rate (R_r), which is actually of more interest, because it allows off-site dose calculations. R_r is the fraction of the total activity in the soil released per second. This provides a source term for meteorological calculations to determine downwind population exposure. R_f only provides dose or exposure for a person present on the contaminated area, which is somewhat academic for real life situations, because plant personnel only spend limited time on or near the pad field. Our Pu flux data makes it possible to estimate R_r, but estimates of the Pu particle plume profile have to be made.

Before proceeding to estimate R_f and R_r, the limitations for applying these factors should be understood. Resuspension factors/rates ignore the physical parameters affecting resuspension, such as wind speed, vegetative coverage, soil moisture, precipitation and contaminant/host particle size. Also, a good knowledge of the Pu surface distribution is assumed, as well as airborne concentration over the whole area in question. As Sehmel (HA80cp269) correctly points out, realistic prediction of the relationship between surface concentration and airborne concentration is fraught with uncertainties. Such data is very site specific and depends on how the contaminant found its way into the soil/vegetation and how long the contaminant has weathered in. For example, resuspension for the first few weeks, after a tracer was sprayed onto cut grass, was orders of magnitude higher (RE79p27) than our data given below. Our preference is to use actual Pu concentration data and then draw conclusions.

Sehmel (SE72) probably made the best estimates of maximum resuspension factors at RFP for a special situation in 1969, when Pu releases were high with no vegetation on the field during the pad remedial project that involved earth moving machinery. Samples were taken for time periods as short as six hours in the source area. The R_fs ranged from 10^{-9} to 10^{-5} m^{-1}. However, these factors are no longer applicable, unless similar areas of fresh soil are exposed.

We calculated a R_f range of 10^{-13} to 10^{-10} m^{-1} limited to areas near the pad field sampler (RF4036P44). The variability in soil Pu activity (see Fig. 2) and ground cover, raises

serious questions about generalizing from these values to the whole field. The same applies to the calculations for R_r, which was estimated at 2×10^{-12} sec^{-1} for the whole 903 pad area. This calculation required an estimate of the average air flow over the field and the resulting fetch for resuspended particles. This parameter was used to estimate the total emission of Pu from the field to be 200 uCi per year.

The question of Pu transport to populated areas is better answered by downwind Pu concentration data provided earlier in this report. These long term measurements show that the pad field resuspended Pu does not contribute appreciably to off-site dose. We could not discern the pad field influence beyond about 1.5 km. To further confirm this observation, we suggest future studies involving additional air sampling along Indiana Street with improved air samplers, that do a better job of efficiently collecting larger particles (RF3650p20). The Pu samples should be analyzed for the Pu240/Pu239 ratio as well as samples of nearby soil to identify the sources of the Pu (i.e., fallout vs. RFP Pu).

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PARTICLE RESUSPENSION: A REVIEW

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Published numerical values of resuspension variables, rates, factors, and weathering half-lives are summarized. Results of the review show the great uncertainty in accurately predicting resuspension. Resuspension rates range over six orders of magnitude from 10^{-12} to 10^{-4} fraction/sec, resuspension factors over nine orders of magnitude from 10^{-10} to over 10^{-2} m^{-1}, and weathering half-lives from 35 days to years. Data are yet needed to evaluate resuspension changes as a function of pollutant particle properties, including chemical composition, solubility, and shape.

Introduction

Resuspension has been receiving increasing attention within the last several years, as is evidenced in the Atmosphere-Surface Exchange of Particulate and Gaseous Pollutant (1974) conference proceedings (Engelmann and Sehmel, 1976), which consider both resuspension and dry deposition. Resuspension can be a continuing problem after toxic materials are deposited on environmental surfaces. Once the material is on the ground, it may be recycled into the atmosphere to be inhaled by man.

Accurately predicting resuspension is important in evaluating effects of airborne concentrations arising from surface contamination. Mass transfer across the air-surface interface is important for all toxic materials, regardless of their size. If these materials are respirable, predicting airborne concentrations that might be an inhalation concern is crucial. These airborne concentrations can be calculated from atmospheric diffusion and transport models which include a resuspension rate boundary condition for mass transfer across the air-surface interface. Unfortunately, man is most often exposed to the hazard of particle inhalation in the bottom 1.5 m of the atmosphere, a region which is generally neglected by meteorological models used to simulate the downwind transport and diffusion of pollutants.

Mass-transfer processes reviewed here are both resuspension and suspension. Resuspension describes material deposited from the atmosphere and then subsequently re-entrained or resuspended into the atmosphere. Suspension describes the subsequent insertion of particles into the atmosphere which were originally deposited on a surface by some nonatmospheric process such as an industrial spill. Since the combined processes are usually referred to as resuspension, this term will be used to describe both resuspension and suspension. Another reason for referring to these two processes by one term is that we cannot distinguish between the subsequent behavior of particles deposited from the atmosphere or from an industrial spill. After deposition, most pollutant particles lose their individual identity by becoming attached to host soil particles. When pollutant particles are transported downwind, they are usually attached to host soil particles (Sehmel, 1976b; 1978b) rather than transported as discrete pollution particles.

Usually resuspension concepts are applied to surface contamination rather than bulk soil erosion, but the soil erosion data reported in this review have a general application to resuspension concepts. Prediction of agricultural erosion, and hence the role of soil particles, is a specific resuspension application. The results of agricultural erosion research have been summarized (Skidmore and Woodruff, 1968) in terms of a soil erodibility index. From these studies, predictions of average soil loss in a field, ton/acre/month, can be estimated.

Resuspension rates are needed as boundary conditions for atmospheric transport models. Although resuspension measurements and some correlating bases exist for predicting mass transfer at the air-surface interface, predictive transport models need extensive validation and development. Many models have been developed to describe the combined deposition, resus-

pension, diffusion, transport deposition, and resuspension of an airborne plume. However, as will be shown, insufficient and inadequate data are available to validate these models.

Although data are insufficient to validate predictive transport models, models have been developed to describe mass transfer within the air–surface interface in terms of mass-transfer resistances. Multibox resistance models are often used as a mathematical convenience to simplify the continuum mass-transfer processes including resuspension (Calder, 1961) by utilizing an initial box in the lower 1 mm, and additional boxes for incremental distances above that 1 mm. These models have been developed by Bennett, Hill, and Gates (1973), Bennett and Hill (1975), Caporaloni et al. (1975), Garland (1976), Liss and Slater (1976), Markee, Andrews, and Jubach (1977), Murphy (1976), Sehmel and Hodgson (1978), Slinn (1976a, 1976b, 1977), and Slinn et al. (1978).

In applying multibox resistance models to diffusion and transport models, usually only two boxes can be used, an upper box described by standard meteorological diffusion and a lower box described by either a resuspension rate or a dry deposition velocity. Resuspension rates are needed in these models, and resuspension rate predictions can be based upon the data summarized in this review.

The objective here is to assist meteorological and health physics modellers and users of model predictions by (1) reviewing the resuspension literature, (2) discussing some resuspension variables, (3) summarizing both wind caused and mechanically caused resuspension rates and factors, and (4) summarizing weathering half-times describing airborne concentration decreases with time. Since these data are summarized, modellers are assisted in selecting numerical values required in their model applications.

Results of this review will show that predictive resuspension mass-transfer capabilities do not have a sufficient data base to consider independently both natural and man-generated particles. In addition, results will show that resuspension data are still needed to evaluate changes in mass-transfer behavior caused by particles of different chemical composition, solubility, or particle shape.

A summary of resuspension research has been published by Engelmann and Sehmel (1976). That volume, as well as this review and its references, will probably lead one to most published results on resuspension.

Soil Transport

Direct experimental evidence of surface pollutant removal by wind stresses is still preferable to model predictions since there are limited data for model validation. Nevertheless, model predictions could be made based upon the wind stress physics acting upon individual sand grains. That is, wind resuspension rates are the integral over wind stress effects on a microscale. Wind stresses on individual sand grains have been estimated by several investigators through sand grain models. In these modelling studies, force balances including frictional drag, form drag, gravitation, and the Bernoulli effect have been considered (Akiba, 1975; Andres, 1972; Chepil, 1950a; 1959; 1965; Ford, 1957; Kawamura, 1975; Malina, 1941; Slinn, 1976b). These theoretical models have limited applicability to resuspension physics over a wide surface area since model results require integration over the entire surface area, a procedure involving formidable uncertainties. In addition, models of surface forces on individual particles need some means by which to describe the effects of avalanching probabilistically. These models will not be described further in this review.

Nevertheless, modes of pollutant resuspension transport should be similar to soil transport movements. These soil movements can occur on several scales. On a small scale, many sand grains tend to move simultaneously in a series of intermittent bursts confined to small surface areas. After movement ceases, the area remains quiescent until another short burst occurs (Sutherland, 1967). As wind speed increases, particle bursts become more active until particles are suspended and transported into the air.

Larger-scale erosion is described by three terms, which express continual soil movement as a function of surface stresses: saltation, surface creep, and suspension. Differences in motion between these three are gradual. The transport mode depends on particle diameter, wind speed, and wind turbulence. Saltation describes particles, ranging in diameter from about 100 to 500 μm (Newman et al., 1976) that jump or bounce within a layer close to the air–surface interface. Particles transported by surface creep range in diameter from about 500 to 1000 μm. These surface creep particles move by a sliding or rolling motion and are pushed along the ground by wind stresses and momentum exchange from the impact of smaller particles transported in saltation. Particles smaller than about 100 μm in diameter move by suspension and tend to follow air motions. The amount of soil eroded by saltation, surface creep, and suspension varies greatly for different soils (Chepil, 1965; Oksza-Chocimowoski, 1976): suspension moves from 3% to 40% by weight, saltation 50% to 75%, and surface creep 5% to 25%.

Saltation

Of the three types of large-scale erosion described above, saltation (Chepil, 1945a) moves the greatest amount of soil. Saltating particles rotate at speeds from about 200 to 1000 rev/sec and are ejected almost vertically into the airstream. The velocity of this vertical ejection is comparable to the friction velocity, u_* (Owen, 1964). While gaining considerable horizontal

momentum from the wind, saltating particles descend with rather flat trajectories—at an angle of between 6° and 12° from horizontal—to strike the soil surface and thereby cause avalanching. After striking the surface, saltating particles either saltate again or cause surface creep. In either case, avalanching can cause some particles to move in true suspension.

Suspension

Suspension describes the movement of soil particles below about 100 μm in diameter. Suspension transport may be described by meteorological diffusion models. Suspension particles are lifted by the wind when saltation particles strike the surface. If it were not for saltation movement, suspension particles would not be eroded readily. Dust less than 50 μm and especially less than 10 μm in diameter is extremely resistant (Chepil, 1943) to wind erosion. However, when suspension particles are mixed with coarser particles capable of saltation, they are easily transported by suspension.

Threshold wind speed

The wind speed necessary to begin soil erosion, termed the threshold speed, depends upon a complicated set of factors. One factor is whether a surface crust has formed on an exposed field (Chepil, 1945b). The wind speed required to initiate soil erosion in any given season is determined in part by this crust. The erosion of soil with a surface crust can be initiated by wind speeds of 6 to 13 m/sec at an elevation of 0.3 m. Once that crust is broken in a bare field, wind speeds greater than the lower limit of 6 m/sec at 0.3 m will continue wind erosion.

The smallest threshold speeds occur for particles ranging in diameter from 100 to 150 μm. For smaller particle diameters, the threshold speed increases with a decrease in particle size. For particle diameters larger than 150 μm, the threshold speed increases. For mixtures of soil particle sizes, the threshold speed is smaller than that required to erode only the largest particle. In other words, the threshold speed increases directly with average particle size and decreases with a wider range of particle diameters in the soil (Chepil, 1943). Bagnold (1941) indicates particles above approximately 100 μm will saltate and be carried aloft only if the wind speed becomes significantly greater than the threshold speed.

Horizontal soil fluxes

The results of soil erosion rates are important to resuspension studies since pollutants are often transported on eroding soil particles. The results of erosion research are often reported in terms of a horizontal, soil particle flux with particular emphasis on the transport of larger particles. O'Brien and Rindlaub (1936) developed the original relationship between the amount of sand transported and wind speed. These measurements

were made at Clatsop Beach at the mouth of the Columbia River. At this location, the sand has a mean diameter of about 200 μm. Their equation has been adjusted to the amount of sand drift, Q, in kg/m width/h. Results show

$$Q = 0.79(u^3) \qquad (1)$$

for wind speeds (u in m/sec) greater than 6 m/sec at an elevation of 1.5 m. Bagnold (1941) found similar results for the movement of sand during dust storms in the Egyptian desert. For a mean sand particle diameter of 250 μm, the amount of sand transported is expressed by the relationship

$$Q = 5.2 \times 10^5(u - u_T)^3, \qquad (2)$$

where u is the wind speed in m/sec at 1 m height, and u_T is the threshold speed equal to 4 m/sec. By comparing coefficients, it is obvious that horizontal fluxes are site specific.

Both expressions show that the horizontal flux of sand is proportional to wind speed, u^3. Similar results are reported in the wind erosion equation developed by Chepil (1951). In wind tunnel experiments, the soil movement rate was a function of the friction velocity, u_*^2. However, wind tunnel results indicated the air capacity to transport soil, i.e., the total horizontal flux of sand, was a function of the friction velocity, u_*^3.

Hsu (1971) developed a correlation for predicting sand transport as a function of mean sand particle diameter for data published between 1936 and 1963. In this correlation, the sand transport rate is also proportional to u^3.

Resuspension Coefficients

Modellers can use either resuspension factors or resuspension rates to predict the airborne effects from resuspension. Resuspension was described in the early literature (Langham, 1971) by a resuspension factor, RF, which is the ratio of airborne contaminant concentration per unit air volume, χ, divided by the contaminant surface concentration per unit area on the ground, G:

$$RF \text{ (units of } M^{-1}) = \frac{\chi, \text{ airborne concentration } (M^{-3})}{G, \text{ surface concentration } (M^{-2})}. \qquad (3)$$

The resuspension factor cannot be predicted accurately. The magnitude of the resuspension factor, which has ranged in experimental studies from about 10^{-10} to 10^{-2} m^{-1} (9×10^{-11} to 3×10^{-4} m^{-1} for wind resuspension and 1×10^{-10} to 4×10^{-2} m^{-1} for mechanical stresses from man's activities), depends in

part upon the sampling techniques used to measure particle concentrations. The airborne concentration is measured at some height above the ground surface concentration measurement site. Neither the same height of air sampling nor depth of surface soil sampling is always used although breathing height and a 1-cm-depth sample of the ground might be used. The definition given to "surface concentration" must be precisely stated, since the term can mean anything from the pollutant in the first millimeter-depth in the soil per unit area to that in the first several centimeters-depth per unit area. Similarly, the definition of "airborne concentration" is not often precise because the measurement height is not specified. Obviously, different measurement heights and contaminant particle sizes on the surface will certainly change the resuspension factor.

Some of the prediction uncertainty for the resuspension factor may be caused by several deficiencies in the resuspension factor concept. One deficiency is that the airborne concentration for inhalation is assumed to be related to the surface concentration on the soil at a person's feet rather than upwind. Another is that resuspension factors are applicable only for the conditions for which they were determined. At present, no general method has been devised to extrapolate or predict resuspension factors for general situations. Even taking average resuspension factors is a risky concept because of the orders of magnitude of uncertainty within a single experiment in measured resuspension factors. A third deficiency is that most resuspension factors have not been determined under conditions in which the resuspension source characteristics were carefully controlled. In most experiments the pollutant sources were nonuniformly distributed across an area, and the pollutant source particle size distributions were unknown.

Although resuspension factors can be used to estimate airborne concentrations above a contaminated area, resuspension factors do not describe particulate resuspension rates nor can they realistically be used to predict downwind air exposure and ground deposition. For downwind calculations of air exposure and ground deposition, resuspension rates and airborne particle concentrations as a function of height are needed so that the resuspension coefficient, i.e., the rate, is dimensionally consistent with particle diffusion–deposition models. For use in these models, a group of suspension (resuspension) rates, S, must be evaluated with units of the fraction of source resuspended/unit time. Suspension rates, rather than resuspension rates are usually determined experimentally.

Three soil size variables must be considered, though one may dominate, since soil particle movement occurs by different mechanisms (Bagnold, 1941; 1960) for different sized particles. The total resuspension rate, S, is

$$S = f(S_W, S_M) = f'(S_{air}, S_{salt}, S_{sc}), \qquad (4)$$

where S_W are the wind-caused resuspension rates and S_M are the mechanically caused resuspension rates. In both cases the three modes of soil transport, S_{air}, S_{salt}, and S_{sc} are a function of particle size. The S_{air} is the suspension rate for particles moving in true airborne suspension, S_{salt} is the suspension rate for particles moving in saltation, and S_{sc} is the suspension rate for particles moving in surface creep.

Boundary conditions for atmospheric transport models can be calculated when the contaminant surface concentrations and resuspension rates are known. The upward resuspension flux boundary condition is determined by multiplying the surface concentration by the resuspension rate. Conceptually, this calculation is simple. In reality, the physics are complex. Similar to resuspension factors, a complication arises in the determination of the surface contamination level. To illustrate this complication, assume (1) the resuspension rate was constant and (2) the total surface contamination loss to air was small. Even in this case, resuspension fluxes could change with time since the surface contamination source could also change as a result of contaminants washing, weathering, and penetrating more deeply into the soil. This penetration was investigated by Krey, Hardy, and Toonkel (1977). After time, the contaminant soil concentration decreases exponentially with depth into the soil. Consequently, depending on the soil depth used to define the source strength, widely different resuspension fluxes could be calculated. If the source strength is measured to include all contaminants penetrating to greater depths, the calculated resuspension flux would remain constant. If only the surface contaminant is measured, the calculated resuspension flux would decrease with time.

Conceptually, both resuspension rates and factors are related only to the contaminant that the wind stress can act upon, which might be within the top millimeter or so of a soil surface. However, experimental practices for defining the surface contamination source strength are fraught with experimental problems. To determine the contaminant surface concentration experimentally, the contaminant has been calculated for varying sampling depths. Often the top 1 cm of soil is sampled. This presents a limited interpretation problem if the contaminant is only on the top surface of that 1 cm section. However, if the contaminant is uniformly distributed throughout the top 1 cm, the source strength estimate is too large. In fact, source strengths have been evaluated at even greater depths.

Surface sampling techniques directed toward obtaining contaminant particle size also cause an inconsistency in determining resuspension rates and factors.

Total soil surface sampling distorts resuspension physics because it ignores how an airborne concentration, of a particular particle size, is related to the contaminant surface concentration in that same or different particle-size ranges on the ground surface.

Airborne Concentration Profiles

Rather than discuss transport models* separately, this review discusses only the expected air concentration profiles. These profiles are the airborne pollutant concentrations as a function of height above ground. Figure 1 shows idealized, two-dimensional, airborne pollutant concentrations as a function of height for three different situations. Figure 1A shows airborne concentrations for resuspension from a local source. In this case, airborne concentrations are maximum at ground level and decrease with height. Downwind from that local source, airborne concentrations change, as suggested in Fig. 1B, and show a depletion near ground level as a result of particle deposition. Obviously, large dry deposition rates produce more depletion than low dry deposition rates. For both high and low dry deposition rates, a maximum airborne concentration occurs at some height above ground level.

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*Many meteorological models are used to calculate the downwind transport, diffusion and deposition of particles. In general, these transport, diffusion models can be modified to include the effects of surface mass-transfer including resuspension. Some of these model approaches include Sutton's equations (Chamberlain, 1955), Pasquill's model (1962, 1974), Pasquill's equations (Brook, 1968), a Gaussian plume model using surface depletion (Horst, 1977a; 1977b), Gaussian plume models by others (Amato, 1976; Davis, 1978; Healy, 1974; 1977a; Mills *et al.*, 1974; Scriven and Fisher, 1975; Shreffler, 1975; Travis, 1976; Trevino, 1972; Van der Hoven, 1963), Slinn's theoretical models (1976a, 1977, 1978), and finite difference models (Draxler and Elliott, 1977). In addition, surface soil transport by saltation and surface creep has been considered by Amato (1976). These models will not be specifically described in this review because there is much uncertainty as to which one of them, if any, has been validated.

Airborne pollutant concentration profiles resulting from an upwind surface release are shown in Fig. 1C. For equal total deposition, both a stable and unstable condition are considered. Airborne concentrations at ground level are less for the unstable case since pollutants mix upward more rapidly than in a stable atmosphere.
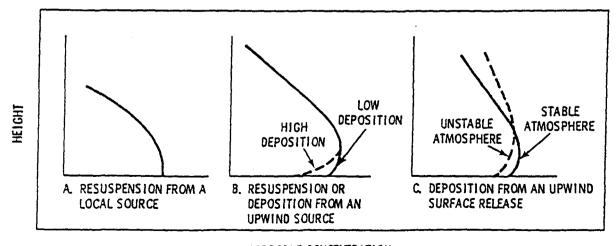
Resuspension Considerations

There are many qualifications in predicting airborne concentrations from resuspension. Despite these uncertainties, much is known about resuspension. Variables that influence resuspension predictions and physics will be considered.

Airborne pollutant particle diameter

In air–surface mass-transfer exchanges, the most important particles to consider for resuspension are those that can be inhaled and potentially cause damage in the lung. In general, the respirability of a particle depends upon its diameter. Particles less than $3.5~\mu m$ in diameter are most often considered respirable, while particles as large as $15~\mu m$ can be inhaled. The sizes of inhaled particles determine where they will be deposited in the tracheal–bronchial tree and the relative hazard of the particles. Smaller particles are deposited in the alveoli; larger particles in the neighborhood of $15~\mu m$ or greater are removed from the air in the nasal passageway.

Soil erosion

Some soil erosion results can be interpreted in terms of resuspension rates. Total soil erosion over thousands of years can be estimated by the depth to which roots or rocks are exposed. In Greenland, the average erosion in exposed areas was 9 to 37 m in depth per

Fig. 1. Idealized airborne pollutant concentrations as a function of height.

thousand years (Judson, 1968). In vegetated areas, surface losses were less, only 3 m per thousand years, and even less in Utah and California where root exposure depths indicate erosion between 0.02 to 0.1 m per thousand years. An estimate for continent-wide erosion is 0.024 m per thousand years. These average surface-depth erosion rates, ranging from 6×10^{-11} to 1×10^{-7} cm/sec, correspond to an average resuspension rate of from 6×10^{-11} to 1×10^{-7} fraction resuspended/sec for the top 1 cm of soil.

Agricultural wind erosion results are reported only in terms of a wind erosion equation (Woodruff and Siddoway, 1965; Skidmore and Woodruff, 1968; Skidmore, 1976), which is a product of a soil erodibility index, a climatic index, a soil ridge roughness factor (Chepil, 1950), the unsheltered field length, and an equivalent quantity of vegetation. The product of these terms is proportional to the annual soil loss, tons per acre or depth loss per unit time. This equation can predict soil losses up to about 340 ton/acre/yr, which corresponds to a predicted maximum average surface loss of about 4 cm/yr or 1×10^{-7} cm/sec. If the soil surface "source" depth is assumed to be 1 cm, the surface loss corresponds to a resuspension rate of 1×10^{-7} fraction of source resuspended/sec.

Tabulated resuspension variables

Variables influencing resuspension are listed in Table 1 under these classifications: particle properties, soil properties, particle–soil interaction, surface properties, topography, and meteorological variables. Some variables in each column will be discussed briefly.

Pollutant particle properties. In the first column of Table 1, one of the most important variables may be particle diameter since respiration and inhalation are functions of particle diameter. Thus, one would like to characterize the particle diameters of the resuspension source as a step in predicting airborne concentrations. However, at the present time, the effects on resuspension of the diameter of deposited pollutant particles cannot be predicted.

The methods employed to determine particle size distributions in collected soil samples can change the pollutant distribution and even the distribution of host soil particles. Changes in pollutant particle attachment, for instance, can be expected during particle sizing with either liquid or air elutriation. In these methods, smaller respirable particles are released from host soil particles. Rosinski et al. (1976) have shown that respirable particles are released when a particle is dropped into liquid. Sehmel (1978a) has shown that respirable particles are released when a soil particle is dropped on a solid surface.

Soil properties. Soil properties influencing particle resuspension are listed in the second column of Table 1. An overriding factor influencing soil erosion is soil moisture content. As long as soil is saturated,

resuspension will not occur; as the surface layer dries, however, resuspension can occur. The increased resuspension rate with decreasing surface soil moisture content is unknown. However, the threshold friction velocity for initiating sand saltation, u_{*T}, was investigated by Belly (1964) as a function of sand moisture content. For conditions of that investigation, the threshold friction velocity was expressed by

$$u_{*T} = A(1.8 + 0.6 \log w) \sqrt{(\sigma - \rho) g d / \rho} , \qquad (5)$$

where A is an adjustable coefficient, approximately equal to 0.1.

After precipitation and drying occur, a surface salt-crust can form. This crust tends to minimize soil erosion until it is broken by mechanical action or by the upheaval of soil caused by thawing and freezing (Caine and Morin, 1976).

Particle–soil interaction. The interactions of pollutant particles to host soil particles are listed in the third column of Table 1. Though measurements of soil–surface interactions have been made (Corn and Stein, 1965; Corn, 1966; Walker and Fish, 1965; Punjrath and Heldman, 1972a; 1972b) in laboratory experiments for very simple surfaces, the results have not been generalized to the more complex interactions in the environment.

Resuspension can occur from vegetation as well as soil. For instance, Aylor (1976) determined that large numbers of particles were resuspended from corn. Similar work has been done by Chamberlain (1970, 1974, 1975), Heinemann, Vogt, and Angeletti (1976), Heinemann et al. (1976), and Peters and Witherspoon (1972).

Topographical variables. Though little is known about the effect of regional topography on resuspension other than visibility differences caused by airborne dust (Orgill and Sehmel, 1976), topography effects may influence average resuspension rates. Most resuspension literature only discusses desert or semiarid areas, rather than humid areas, while soil erosion data apply primarily to agricultural areas.

Little data has been collected to assess the resuspension that can occur in urban topographies. Some data show (Newman et al., 1976) that material is "refloated" by traffic on city streets. These suspended materials are primarily abraded tire and pavement particles, and recycling pollutant particles. Only limited data quantifies the refloatation rate (Sehmel, 1973) (see Figs. 6 and 7 discussed later in the section on *Vehicular traffic induced resuspension*).

Meteorological variables. Though many meteorological variables are assumed to influence resuspension, their effects have, for the most part, not been quantified. The physics of turbulent wind stresses next to the surface are still being studied. In the literature on fluid flow, an undisturbed laminar boundary layer is no

Table 1. Variables which influence resuspension.

Pollutant particle properties	Soil properties	Particle–soil interaction	Surface properties	Topography	Meteorological variables
Chemical reactivity	Moisture content	Attraction forces	Areas of erodible surface	Agricultural	Air density, affected by
Density	Ratio of erodible to non-erodible fractions	• Molecular forces: Van-der-Waals forces	Cohesiveness of particles	Broken	• Humidity
Hydrophobic		• Chemical binding forces	• Binding action of materials	Desert	• Pressure
Hydrophilic		(Valence forces)			
Particle diameter	Soil size frequency distribution	• Electrostatic forces	• Surface moisture	Diurnal	• Temperature
• Respirable	Specific gravity	• Magnetic forces	Cover	Flat	Soil burden carried
• Nonrespirable	Structure	Capillary forces at the surface of aggregates filled with liquid	Obstructions	Forested	• Influence on air velocity
• Frequency distribution	• Organic matter	Chemical reaction	Roughness, large scale	Hills	•Avalanching
Shape	• Lime content	Contact area effect on adhesion	• Mechanical turbulence	Humidity	• Saltation
Surface roughness	• Texture	• Particle shape	• Overall sheltering	Seasonal	Surface moisture content
	Temperature	• Particle size	Roughness, small scale	Temperature	• Dew
		• Nature of particle and surface	• Orientation of obstacles	Undulating	• Frost upheaval
		• Surface roughness	• Percent of area covered by non-erodible aggregates or obstacles	Uniform	Relative humidity
		• Adhesion of agglomerates vs individual particles		Urban	Temperature
		Crystallization of dissolved substances	• Sheltering of individual particles	Valley	• Surface dry
		Deposition of suspended colloidal particle	• Stability of aggregates against abrasion and disintegration by moisture and watering	Water	• Surface drying
		Hardening binders			• Surface frozen
					• Wind speed
		Liquid bridges	Temperature		Wind removal forces
		Mineral bridges	Topographic features		• Mean wind speed
		Relative humidity	• Broken		• Frequency, period, and intensity of gusts
		Temperature	• Uniform		penetrating to the surface
		Time of contact	Vegetative cover		• Transient and steady drag
		Viscous surface coating	• Live vegetation		• Verticle wind profile
			• Plant residue		• Stability
					• Vertical turbulent exchange
					• Surface roughness
					• Temperature stratification
					• Wind direction

longer considered to occur next to a smooth surface. Fluid turbulence penetrates to the surface intermittently and unpredictably, especially over rough surfaces. Turbulence may penetrate in a burst directed toward the ground or in a small dust devil. The author has seen small dust devils, on the order of 10 cm in height and spaced about 1 m apart, resuspend zinc sulfide tracer from an asphalt surface. These dust devils may have been caused by solar heating rather than turbulence from the average air speed.

Resuspension Factors

The resuspension factors reported in this section can be used in estimating airborne concentrations arising from resuspended surface sources. These resuspension

factors were determined from a collection of airborne material sampled without differentiation between respirable and nonrespirable airborne particles.

Wind- and mechanically-induced resuspension

Resuspension can be produced by wind and mechanical disturbance of the soil (Hereim and Ritchie, 1976), but more readily by the latter. Resuspension factors for wind and mechanical resuspension are arranged in graphical form in Fig. 2. Resuspension factors for mechanically caused resuspension are shown in the upper portion of the figure, and resuspension factors for wind stresses are shown in the lower portion. In both cases the data have been ranked in terms of descending order of the maximum resuspension factor reported for each experiment. In addition, the resuspension factor range for each experiment is shown by a horizontal line with vertical end marks. If only a single suspension factor is reported, this datum is shown as an open circle.

Several conclusions can be drawn from this figure. These include the following:

- The figure does not correlate, rather only shows a graphical representation from maximum to minimum reported resuspension factors.
- The range of resuspension factors within some individual experiments can be over several orders of magnitude.
- Mechanically caused resuspension factors range over seven orders of magnitude from 10^{-10} to over 10^{-2} m^{-1}.
- Wind-caused resuspension factors range over seven orders of magnitude from 10^{-10} to 10^{-3} m^{-1}.
- The maximum reported resuspension factor is 10^{-10} m^{-1} for both wind and mechanically caused resuspension stresses.

In utilizing this figure for predictive purposes, I recommend, if possible, locating a similar stress and situation comparable to the one being studied. If this is not possible, I recommend, at least, using a resuspension factor range for sensitivity studies rather than using only a single "average" resuspension factor. Obviously, inspection of this figure shows that our ability to accurately predict airborne concentrations from either mechanically or wind-caused resuspension stresses is extremely poor. Much research is yet needed to adequately understand resuspension processes and to accurately predict airborne concentrations arising from surface contamination.

Airborne concentration half-life

Airborne pollutant concentrations may decrease with time above a resuspension source. This change in airborne concentration with time is reported in terms of

an airborne concentration half-life, the apparent time required for the average airborne concentration to reduce to one-half the original value. This length of time depends at least upon average wind speeds and varies according to the availability of surface contamination to resuspension. Although originally assumed to be constant, the half-life is now considered to change with time.

Half-lives summarized in Table 2 range from 35 days to years. This variability in the half-lives of even the same source material is probably caused by differences in conditions such as wind speed, vegetative cover, time since deposition, and sampling technique at each test site. Some of these half-life data have been combined into overall models (Anspaugh et al., 1974; Kathren, 1968; Oksza-Chocimowoski, 1976; 1977) that are consistent with Shreve's (1958) original observation that the half-life decreases with time. These models prescribe half-life changes with time.

However, half-lives may not always decrease with time. Half-lives for airborne concentrations at the Plumbbob nuclear test site were originally reported by Wilson, Thomas, and Stannard (1960) and Langham (1971) to range from 35 to 40 days. More recent results indicate a great uncertainty about these half-lives. In the original study starting in May 1957, the 35-day half-life was determined for an air sampling time period of less than 5 months. Subsequently in the summer of 1958, Olafson and Larson (1961) measured airborne concentrations intermittently for times up to 15 months. In comparing the results from these two studies, Healy (1974) found that during the 15-month time period, no appreciable change was reported in airborne concentrations within the data scatter at a site 230 m from the original test location. This result could indicate that resuspension was occurring at an average rate continuously during that 15-month period if the source were uniform with distance. At a site 760 m from the test location, airborne concentrations decreased by a factor of only 1 to 10. These changes in airborne concentration may, therefore, depend upon proximity to the source and possibly local surface contamination levels.

Although the half-life concept appears questionable from experimental measurements of airborne concentrations, there is reason to expect the resuspension source availability to decrease with time. Surface contamination will become attached to soil particles and migrate into the soil structure with time (Krey et al., 1977). In addition, coverage by vegetation and soil from upwind erosion would make the source less available for resuspension. Although less availability with time is suggested, the possibility also exists that the surface contamination that does not migrate could become more readily available for resuspension by degradation processes, possibly by biodegradation (Wildung and Garland, 1977) and solubility changes.
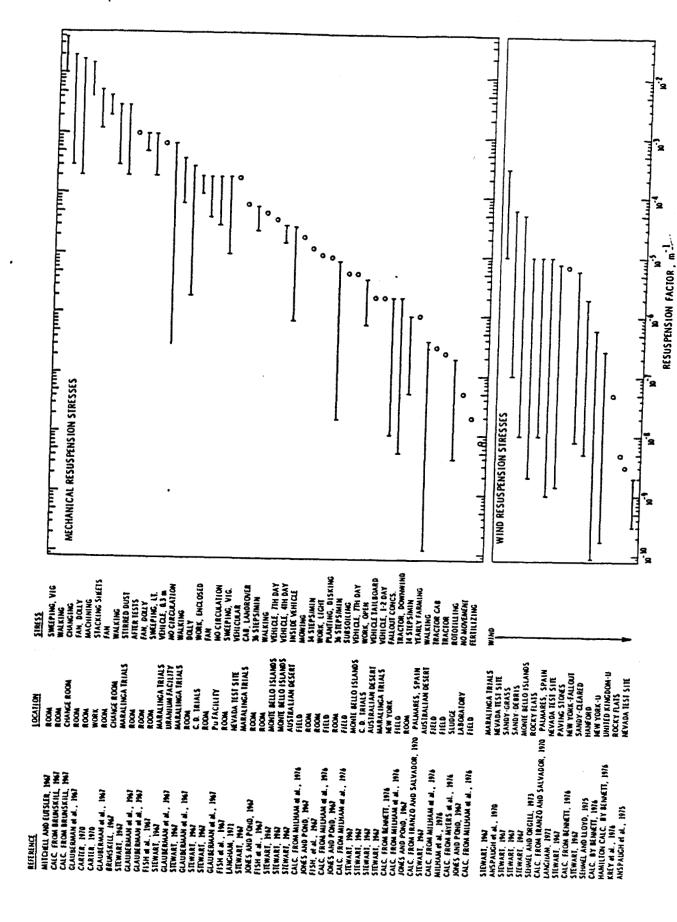
Fig. 2. Resuspension factor ranges from mechanical and wind resuspension stresses.

Table 2. Half-life times for decrease of airborne concentrations near resuspension sites.

Location	Source material	Duration of experiment, months	Half-life for airborne concentration	Half-life for retention on foliage, days	Reference
Hanford	Inert tracer	4 years	Resuspension rates show no continual decrease with years, but seasonal variations. Maximum resuspension rates are in autumn		Schmel (1980)
Nevada test site, Yucca Flat			Concentrations increased from 1956 to 1958		Mork (1970)
Nevada test site Plumbbob			Concentration decreases with time		Shreve (1958)
Nevada test site	Plutonium	15	No concentration change at 230 m distance.		Healy (1974) comparison of Wilson, Thomas
		From 5 to 15	Concentration decreased by only a factor of 1 to 10 at 760-m distance		and Stannard (1960) and Olafson and Larson (1961)
Russia	Alpha and Beta	2 to 4 years	Concentrations decreased 2 to 3 orders of magnitude		Izrael (1971)
		4 years	Practically constant		
Nevada test site, Plumbbob	Plutonium	1 to 5	35 days		Wilson, Thomas, and Stannard (1960)
Nevade test site	Plutonium		35 days		Langham (1971)
Nevada test site, Schooner	^{181}W	1.8	38 days		Anspaugh et al. (1973)
Nevada test site, Baneberry	Gross gamma	9	76 days		Anspaugh et al. (1973)
Hanford Area aged source	^{137}Cs, ^{238}Pu, ^{239}Pu	33	5 months to years		Schmel (1977a, 1977b)
Hanford	Inert tracer	8	8 months		Schmel (1977a)
Rocky Flats after soil disturbance	Plutonium		9 months		Schmel and Orgill (1974)
Rocky Flats	Plutonium	5 years	1 to 2 years		Volchock, Schonberg, and Toonkel (1977)
Field test	Solutions ^{85}Sr, ^{51}Cr, ^{214}Pb and 1 μm diam polysterene			49 in winter 19 in summer (after 7 weeks)	Chadwick and Chamberlain (1970)

The half-life for airborne concentrations is also related to the changes in contamination on vegetation surfaces as a function of time, as shown by experiments measuring the changes in surface contamination levels on plant foliage. Chadwick and Chamberlain (1970), for example, reported a difference in the half-lives of pollutants retained on foliage between winter and summer months. As shown in Table 2, the half-life was greater during the winter.

Resuspension Rates

Resuspension rates are the desired boundary conditions for atmospheric transport and diffusion models.

Experimental resuspension rates will be summarized in this section to show the range of reported numerical values. Resuspension rates have been measured using controlled tracer sources and estimated using sources of opportunity and soil. Sehmel (1977a, 1980) experimentally measured resuspension rates using tracer particles. In these experiments, resuspension rates were measured as a function of wind speed as well as respirable and nonrespirable particle diameters. Mechanical resuspension was measured for vehicular traffic on asphalt and in cheatgrass areas and for pedestrian traffic on asphalt. Sehmel's use of mass balance techniques to determine resuspension rates is a more direct measurement method than in early tracer resuspension data reported by Healy (1955, 1977b) and Healy and Fuquay (1959, 1959). In that early research, airborne concentrations of suspended tracers were estimated using air samplers located only at ground level. Later, a diffusion model was used to calculate apparent resuspension rates.

Wind-induced resuspension

Resuspension rates cannot be reliably predicted as a function of the diameters of the source particles or the physical and chemical properties of the particles. There are too few data even to predict differences between resuspension rates for the same pollutant for surfaces with different vegetation coverage. Airborne concentrations produced by wind stresses have been reported as a function of both wind speed and friction velocity. Resuspension data summarized in Table 3 show that air concentrations increase from the 1.1 to the 6.4 power of wind speed or friction velocity. Therefore, the dependence of particulate airborne concentrations on wind speed is uncertain.

The particle resuspension rates summarized in Table 4 have either been calculated using transport and diffusion models in interpreting measured concentrations at only one height or have been measured by the integration of airborne concentrations as a function of height. Though the data determined from concentrations measured as a function of height with controlled resuspension sources is least questionable, resuspension rates for both experimental techniques range from about 10^{-12} to 10^{-4} fraction of source resuspended/sec. This eight-magnitude uncertainty is slightly less than the eleven orders of magnitude uncertainty for resuspension factors.

Average wind-induced resuspension rates for tracer particles were measured (Sehmel, 1977a; Sehmel and

Table 3. Air concentration dependency on wind speed.

Location	Source material	Concentration increases with windspeed to power n u^a	u_*^a	Reference
Nevada test site, GMX	Plutonium		2.2	Anspaugh *et al.* (1976)
Nevada test site, Yucca Flat	Plutonium	Concentration increases a factor of 10 from $u < 2$ m/sec to $u =$ 5 to 7 m/sec		Mork (1970)
Rocky Flats	Plutonium	2.1		Sehmel and Orgill (1973b)
Rocky Flats	Plutonium	5.9		Sehmel and Lloyd (1976c)
Hanford "Onsite"	Plutonium	1.0 to 1.5		Sehmel (1977b)
"Offsite"	Plutonium $u < 5$ m/sec $u > 5$ m/sec	1.1 3.0 to 9.3		Sehmel (1977a)
Hanford	Tracer	1.0 to 4.8		Sehmel (1977a)
Hanford	Tracer	Two sites: 3.4 avg. and 3.5 to 13.8		Sehmel (1980)
Eroding field	Soil particle diameter, μm 0.4 to 0.6 0.6 to 0.7 2.7 to 2.9		2.6 to 3.4 5.0 6.4	Sehmel and Richmond (1978d)
Eroding agricultural field	Soil		6.38	Shinn *et al.* (1976); Anspaugh, Shinn and Phelps (1975)
Nevada test site desert pavement			2.1	

Table 4. Resuspension rates.

Location	Source material	Resuspension rate, sec^{-1}, determined from		Reference
		Calculation using transport model and concentration at only one height	Integration of measured concentrations	
Corn leaves	Pollen		10^{-6} to 10^{-4}	Calc. from Aylor (1976)
Hanford	Tracer		10^{-13} to 10^{-7}	Sehmel (1980)
Nevada test site	Plutonium	2.7×10^{-12} to 4.8 to 10^{-10}		Anspaugh, Shinn, and Phelps (1975a)
Nevada test site, GMX	Plutonium	10^{-12} to 10^{-10}		Healy (1974)
Nevada test site	Plutonium	1×10^{-10} to 6×10^{-8}		Healy (1974) calc. from Wilson, Thomas, and Stannard (1960)
Hanford site	ZnS	1×10^{-8} to 3.5×10^{-6}		Healy (1974) recalc. of Healy (1955) and Healy and Fuquay (1958)
Eroding field assumed rate, source is top 1 cm depth	Soil			Erosion rates calc. from Skidmore and Woodruff (1968)
0.5 ton/acre/month		2×10^{-9}		
40 ton/acre/month		2×10^{-7}		
310 ton/acre/month		1×10^{-6}		

Lloyd, 1975) for resuspension from three surfaces, i.e., an asphalt surface, a cheatgrass areas, and an area lightly vegetated with cheatgrass and sagebrush. For average wind speeds of 1 to 4 m/sec, wind resuspension rates for the asphalt surface ranged from 5×10^{-9} to 6×10^{-8} fraction resuspended/sec. For average wind speeds of 1 to 5 m/sec at the cheatgrass area, wind resuspension rates ranged from 5×10^{-9} to 6×10^{-8} fraction resuspended/sec.

Resuspension rates of tracer particles from two lightly vegetated portions of the Hanford area are shown in Figs. 3 and 4 (Sehmel, 1980). These rates are for submicrometer and 6 μm particles as deposited. For the submicrometer tracer site, resuspension rates are shown in Fig. 3 for wind speed increments after March 1974 of 4.5 to 5.4, 5.4 to 6.7, and 6.7 to 9.4 m/sec. In general, resuspension rates at each wind speed remained constant over the total time period. However, there is an apparent seasonal change in resuspension rates. The seasonal variation is not explicit since each experimental time period was determined in part by the occurrence of sufficient desired winds during that particular season. Nevertheless, resuspension rates appear to be higher during the autumn than in the spring and summer.

Most resuspension rates were calculated from total airborne samples. To determine the particle size dependency, each stage of cascade particle impactor samples were analyzed separately for tracer content. For both cases, resuspension rates were calculated assuming the tracer source strength was independent of particle diameter. Resuspension rates indicate that the tracer was resuspended or was attached to all host particle sizes characterized by sampling with cascade particle impactors. In addition, the tracer continued to be resuspended attached to all particle sizes throughout the duration of the experiment.

A resuspension rate wind-speed dependency was calculated from the total airborne samples. For this calculation, each resuspension rate was normalized to the respective resuspension rate measured for the 4.5 to 5.4 m/sec wind speed increment. Using this procedure, a least squares fit of the data indicates resuspension rates increased with wind speed raised to the 3.4 power.

For the 6 μm tracer source, resuspension rates are shown in Fig. 4 for wind speed increments of 3 to 5, 5 to 7, and 7 to 11 m/sec. Resuspension rates were calculated from total airborne samples and also as a function of the collection site within cascade particle impactors. Resuspension rates increased with wind
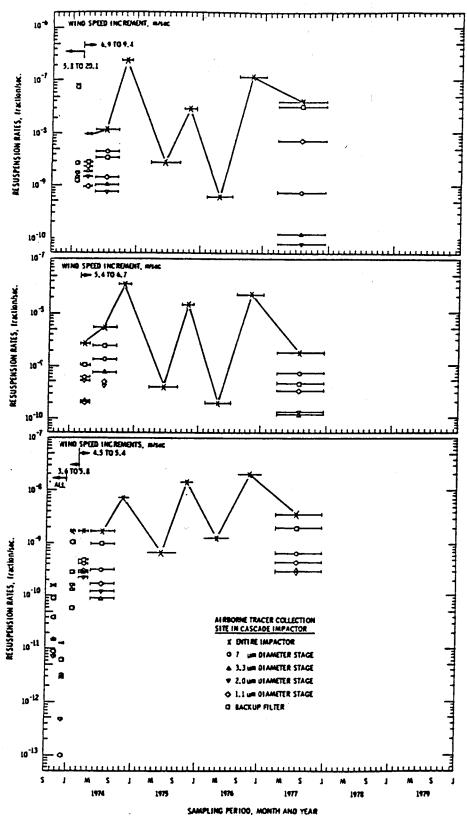
Fig. 3. Resuspension rates as a function of year, wind speed increment, and apparent airborne particle diameter at the submicrometer tracer site.
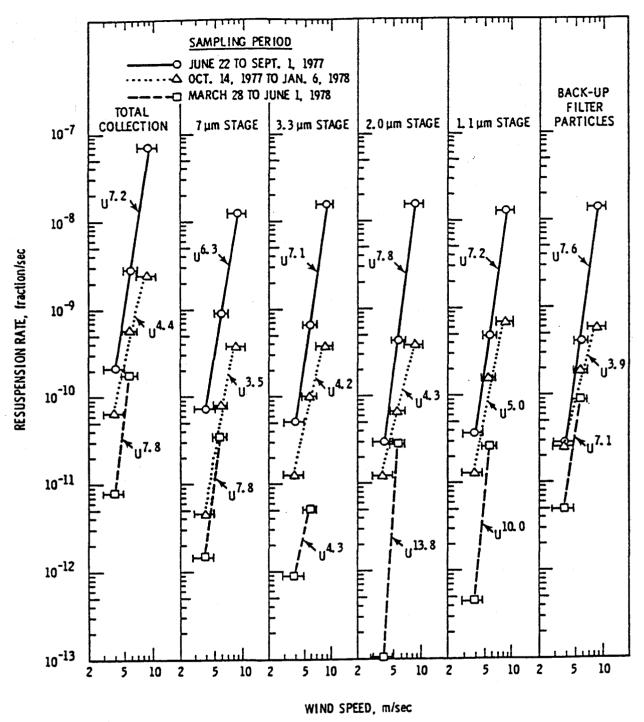
Fig. 4. Tracer resuspension rates as a function of particle diameter, wind speed increments, and average wind speed, U, at the 6 μm site.

speed raised to the 3.5 to 13.8 power. For the time period shown, resuspension rates were decreasing with time.

Average wind-induced tracer resuspension rates of both respirable and nonrespirable particles are shown over lightly vegetated desert in Fig. 5. Average resuspension rates for nonrespirable particles were nearly independent of time and were on the order of 10^{-11} fraction resuspended/sec. Average resuspension rates

for respirable particles ranged from about 10^{-11} to 10^{-7} fraction resuspended/sec and did not decrease during the measurement period.

Correlation for wind-induced resuspension rates

Resuspension rates measured in several experiments (Sehmel, 1975) are plotted in Fig. 6 as a function of aerodynamic surface roughness height. In this figure, resuspension rates range over seven orders of magni-
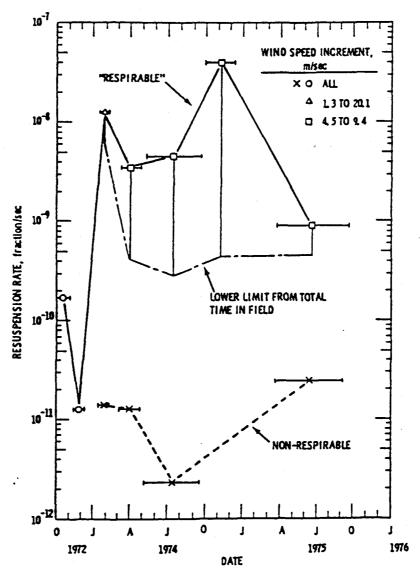
Fig. 5. "Average" wind-caused tracer resuspension rates (lightly vegetated desert on Hanford Reservation).

tude from 10^{-10} to 10^{-3} fraction resuspended/sec. To estimate the fractional source strength removal per year, these rates are multiplied by the 3.2×10^7 sec in a year, i.e., 10^{-4} to 10^{-3} fraction resuspended/yr. Figure 6 is a guideline for estimating resuspension rates for other aerodynamic roughness heights, as illustrated in Table 5. However, this correlation with aerodynamic surface roughness should be used with extreme caution. One would expect resuspension rates are also a function of additional parameters other than aerodynamic surface roughness alone.

Vehicular traffic induced resuspension

Resuspension rates for vehicular traffic on an asphalt road are shown in Fig. 7 as a function of vehicle speed (Sehmel, 1973). The mass-median tracer diameter was 8 μm and all particles were less than 25 μm diameter. Resuspension rates were calculated by

determining the fraction of the tracer resuspended each time the vehicle was driven along the road. The fraction of tracer resuspended each time the vehicle passed ranged from 10^{-5} to 10^{-2}. Resuspension rates increased with vehicle speed. Also, when vehicles were driven on the lane containing the tracer, the resuspension rates were greater than when the vehicle was driven on the lane adjacent to the tracer lane. For vehicle speeds above 20 mph on the tracer lane, resuspension rates for both car and truck passage were found to be comparable.

These rates were also found to depend upon the length of time the tracer particles were on the asphalt road. After the tracer was on the road for four days, particle resuspension rates decreased, as Fig. 8 shows. As before, resuspension rates were greater when the vehicle was driven through the tracer lane than when it was driven on the adjacent lane.
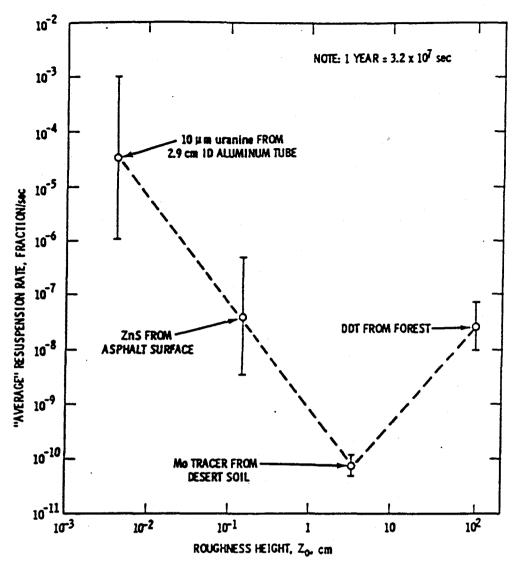
Fig. 6. Initial correlation of wind-caused tracer resuspension rates.

Although increased resuspension rates are expected at higher speeds because of turbulence, Sehmel found that resuspension rates decreased for the cheatgrass road as truck speed increased from 5 to 30 mph (Fig. 9). This decrease can be attributed to the sequence of truck speeds used in the experiment. The initial resuspension experiment was conducted at 5 mph. At this low speed, the most readily resuspended tracer particles were removed from the cheatgrass. Only when the truck speed was increased from 30 to 40 mph, and air turbulence increased as a result, did resuspension rates increase.

Pedestrian induced resuspension

Resuspension rates caused by pedestrian traffic were determined as the fraction of tracer particles resuspended each time a person walked the length of a tracer lane. A man walking along a tracer lane on an asphalt road caused a fraction from 1×10^{-5} to 7×10^{-4} of the tracer to be resuspended with each pass.

Conclusions

This review has demonstrated that resuspension physics are so poorly defined that much research is needed. One problem is that resuspension coefficients have not been adequately defined. The resuspension factor relates airborne concentrations to local surface-contamination levels, but describes neither the vertical flux from resuspension nor the total downwind flux. Therefore, any calculation using an "average" resuspension factor must be qualified as being uncertain within 2 to 3 orders of magnitude because resuspension factors have ranged 2 to 3 orders of magnitude even in a single field experiment.

The resuspension rate describes the fraction of source resuspended per unit time. If the surface contamination level is known, the vertical flux from resuspension is the product of resuspension rates and source concentrations. Resuspension rates measured for both wind and mechanical stresses indicate that mechanical
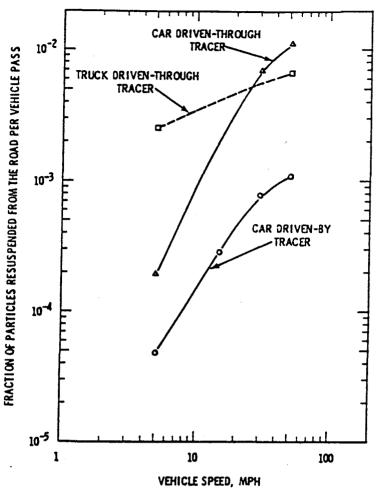
Fig. 7. Rates of tracer particle resuspension caused by vehicle passage over an asphalt road.

Table 5. Typical aerodynamic surface roughness lengths.

Surface	z_0 cm [*]
Smooth mud flats, ice	0.001
Smooth snow on short grass	0.005
Smooth sea	0.02
Level desert	0.03
Snow surface, lawn to 1 cm	0.1
Mown grass	
1.5 cm	0.2
3.0 cm	0.7
4.5 cm[**]	
$u_2 = 2$ m/sec	2.4
$u_2 = 6-8$ m/sec	1.7
to 5 cm	1 to 2
to 60 cm	4 to 9
Long grass	
60 to 70 cm[**]	
$u_{2m} = 1.5$ m/sec	9.0
$u_{2m} = 3.5$ m/sec	6.1
$u_{2m} = 6.2$ m/sec	3.7
Fully grown root crops	14

[*]Based on Priestley (1959) and Slade (1968).
[**]Wind speed at 2 m height.

stresses can resuspend more material at one time than wind, but that effect is short-lived because mechanical stresses usually last over less time and space.

Resuspension research results to date show no significant differences for pollutants from both nuclear and non-nuclear energy sources. Differences cannot be distinguished as a function of chemical properties or particle size.

Experimental resuspension research is needed. In future studies, the experimental conditions must be controlled and defined. Experimental model validation is the crux and requirement for future research of air–surface mass-transfer processes.

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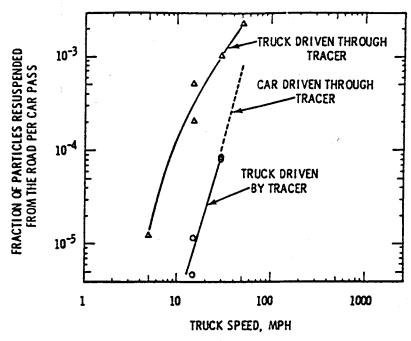
Fig. 8. Rates of tracer particle resuspension caused by vehicle passage over an asphalt road four days after particle deposition.
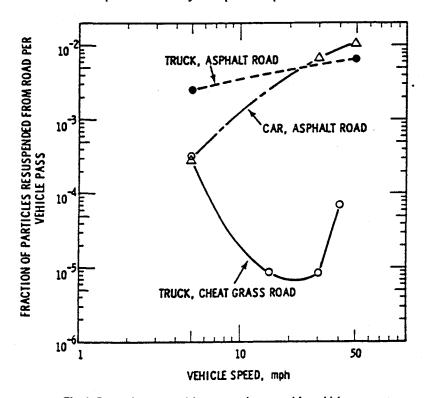
Fig. 9. Rates of tracer particle resuspension caused by vehicle passage over asphalt and cheat grass roads.

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NOTES

A Relationship Between Plutonium Activity Densities of Airborne and Surface Soils*

Introduction

AIRBORNE plutonium concentrations from wind resuspension can either be directly measured or predicted (Se80b). Since costs of extensive direct airborne plutonium concentration measurements may be prohibitive in some instances, model improvements are desirable for predicting airborne plutonium concentrations for environmental impact assessments. Predictive improvements follow for the model based upon the airborne-soil mass-loading calculational approach.

Airborne concentrations from resuspension have been predicted based on considerations of airborne mass loading and pollutant concentrations on the airborne soil (An74; An75). The mass loading approach was developed based on an assumption of equality, i.e. the pollutant concentration per unit mass of airborne soil (A_{air}), is identically equal to the pollutant concentration per unit mass on the local soil surface ($A_{SURFACE}$). In this case, airborne pollutant concentrations are predicted from the equation

$$C = (ML)(A_{AIR}) \qquad (1)$$

where ML = airborne soil mass loading per unit volume of air, and A_{AIR} = pollutant concentration per unit mass of airborne soil. The method was based upon data for plutonium. In this case, the pollutant concentrations on airborne and surface soils are activity densities, A_{AIR} and $A_{SURFACE}$, fCi/g. In developing this method, predicted airborne concentrations, $\mu Ci/cm^3$, were based upon an assumed airborne mass loading of 100 $\mu g/m^3$ and an equality of A_{AIR} and $A_{surface}$. The activity density on airborne solids was not determined.

In using the mass loading calculational approach for predictions of airborne pollutant concentrations, caveats are inherent. These include:

● The pollutant concentration on surface soils may be nonuniformly distributed spatially and as a function of surface-soil particle size distributions.

● The pollutant distributions spatially and as a function of the surface soil particle size distribution are usually unknown.

● The pollutant distributions are known with even less certainty as a function of surface soil particle size distributions as compared to the pollutant distributions on airborne particle size distributions.

● Airborne soils are usually mixtures of soil resuspended from the contaminated surfaces under investigation and soils resuspended and transported from upwind.

The purpose here is to summarize data for plutonium as a pollutant on airborne and surface soils, if both are available at study sites, and to examine the data for relationships between plutonium concentrations on airborne soils and on surface soils near the airborne particulate sampling sites. In practice, surface soil samples are actually soil samples taken to a sampling depth. Only data for sites will be summarized for which the plutonium concentrations on both airborne and surface soils have been investigated. These sites include the Bikini Atoll (Sh80), the Hanford Site (Se80a; Se81) and Rocky Flats (Kr76; Se80a). The plutonium concentrations are reported as activity density ranges, fCi/g of collected soil, i.e. 10^{-15} Ci/g. All plutonium activity density data on surface soils were determined for total soil samples rather than as a function of host soil particle diameters to which the plutonium was attached. Thus, the reported plutonium activity densities are for both respirable and nonrespirable size host soil particles. In contrast, most plutonium activity densities for airborne solids were determined for particles which were respirable, or near respirable.

Because there are only limited activity density data, only a preliminary relationship was sought based on reported activity densities for airborne and surface soils. Improved relationships between meteorological, surface and source characteristics may be developed at a later time, at a time when significantly more validation data have been collected. Until that time, the activity density relationship to be developed significantly improves our ability to predict airborne pollutant concentrations attributable to resuspension.

Activity Density Relationship

Plutonium activity densities of airborne, A_{AIR}, and surface soils, $A_{SURFACE}$, which follow are summarized as ranges measured at Bikini Atoll, Hanford and Rocky Flats. However for both soil sampling convenience and individual program goals, the surface-soil sampling depths in this summary were always greater than the wind resuspension source-depth. Indeed, these sampling depths may tend to reduce surface-soil resuspension-source activity densities in comparison to measured activity densities of airborne soils. Although further refinements in soil sampling techniques is desirable for resuspension investigations, the data summarized are the current data base.

The data for airborne and surface samples were collected in independent investigations. Activity density ranges for airborne soils include the entire data

base (except for one atypical datum point, see Se81) while ranges for surface soils are based upon selected soil sampling sites. Soil sampling sites were those in closest proximity to each sampling site for airborne solids. Distances between selected sampling sites reflect spatial areas at Bikini Atoll, Hanford and Rocky Flats. Distances between sampling sites for airborne and surface soils are greater at Hanford than at Bikini Atoll and Rocky Flats.

A conservative relationship was sought between the experimental activity densities of airborne soils and surface soils, i.e. the maximum observed activity densities of airborne soils as a function of the minimum activity densities on surface soils. Once the relationship is established, conservative (high values) airborne concentrations, $\mu Ci/m^3$, can be calculated. The relationship is shown in Fig. 1.

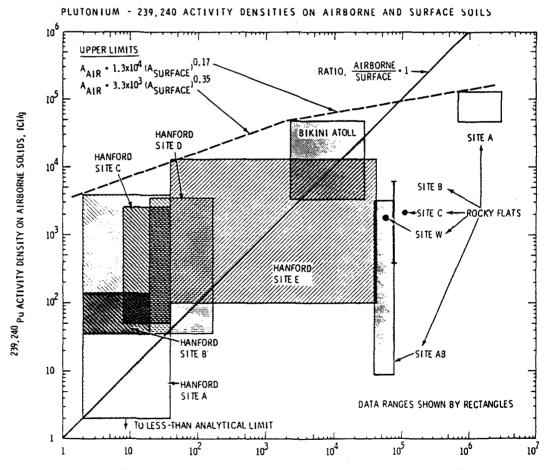Activity density ranges, (A_{AIR}, and $A_{SURFACE}$), are

PLUTONIUM - 239,240 ACTIVITY DENSITIES ON AIRBORNE AND SURFACE SOILS

FIG. 1. Plutonium-239–240 activity densities on airborne and surface soils.

shown in Fig. 1 as shaded rectangular areas. Activity densities of airborne soils range from 10^0 to 10^5 fCi/g, while activity densities of surface soils range from 10^0 to 10^6 fCi/g. The shaded areas intersect and lie on either side of the 45° line representing an equality of A_{AIR} to $A_{SURFACE}$.

The lines labeled "upper limit" represent the conservative estimates sought. In this figure, upper limits are shown by two straight lines for the maximum activity densities of airborne soils. Although these straight lines intersect at an activity density for surface soils of 2×10^3 fCi/g, the intersection is considered to be without further significance other than a mathematical representation of the upper limit data. For smaller activity densities on surface soils, the upper limit is described by

$$A_{AIR} = 3.3 \times 10^3 (A_{SURFACE})^{0.35}. \qquad (2)$$

For larger activity densities on surface soils greater than at the intersection, the upper limit is described by

$$A_{AIR} = 1.3 \times 10^4 (A_{SURFACE})^{0.17}. \qquad (3)$$

These maximum activity lines reflect the presence in collected airborne samples of plutonium and particles from both upwind and resuspended locally. The relative amounts, as exhibited by the data, are a function of the activity density on surface soils. The effects of simultaneous collection of airborne soil particles resuspended from upwind and locally from a resuspension source may be especially evident in the data for Rocky Flats. These Rocky Flats surface soil samples have the largest reported plutonium activity densities.

For activity densities on surface soils less than about 9×10^4 fCi/g, the maximum observed activity densities on airborne soils are greater than on surface soils. The increase reflects several considerations:

• Surface soils contain both respirable- and nonrespirable-sized particles.

• Plutonium activity densities on respirable-sized particles are greater than on nonrespirable-sized particles.

• The observed activity densities are "averages" of activity densities for particles transported from upwind and resuspended locally.

• The relative contributions from upwind and local sources depend upon plutonium particle, surface and meteorological parameters.

For activity densities greater then about 9×10^4 fCi/g, the maximum observed activity densities on airborne soils are less than on surface soils. The decrease reflects the same considerations as just enumerated. The principal difference, in this case, is that soils transported from upwind may contain less contamination than soils resuspended locally.

Airborne Plutonium Concentrations: Source Strength Modified Mass Loading Approach

In the absence of measured annual average airborne plutonium concentrations, conservative estimates of airborne plutonium concentrations from resuspension will be made using a modified mass-loading calculational procedure. The modification entails the use of the upper limit lines from Fig. 1 to relate activity densities of airborne soils to activity densities of surface soils. For the estimation of airborne plutonium concentrations, data are also needed for activity densities of surface soils and airborne mass loadings. Both will be assumed for an example calculation. Based upon reported data, i.e. the range shown in Fig. 1, the activity density on surface soils might be over six orders of magnitude from 2 to 3×10^6 fCi/g. This uncertainty may be unacceptable, except for the fact that the corresponding maximum predicted activity-density uncertainty on airborne soils is less than two orders of magnitude, 4×10^3–1.3×10^5 fCi/g (see Fig. 1). Thus, activity densities of 4×10^3–1.3×10^5 fCi/g of airborne soils will be used for estimating airborne plutonium concentrations.

Airborne soil mass loadings are also needed for estimating airborne plutonium concentrations. Maximum values are sought which are considered realistic. Realistic in this case means mass loadings estimated on an annual basis, a basis considered in evaluating inhalation concerns from airborne radionuclides. In most cases, the annual-average mass loading would be between 10 and 260 $\mu g/m^3$, i.e. somewhere between mass loadings for unpolluted air during low wind conditions and an upper limit corresponding to the 24-hr maximum National Ambient Air Quality Standard (NAAQS) of 260 $\mu g/m^3$. In comparison, the primary NAAQS for the annual geometric mean for suspended particulate matter is 75 $\mu g/m^3$. All these suspended particulate matter concentrations range within nearly one order of magnitude, i.e. from 10 to 260 $\mu g/m^3$. In order to be conservative (high predicted values) in predicting airborne plutonium concentrations in the subsequent calculations, an annual average airborne mass loading of 260 $\mu g/m^3$ is assumed. This corresponds to the 24-hr maximum NAAQS rather than the primary NAAQS of 75 $\mu g/m^3$ for the annual average airborne concentration.

A basis has now been established for estimating an annual-average airborne plutonium concentration attributable to wind resuspension, a conservative estimate. These estimates are for annual average concen-

trations, estimates which are expected to be always greater than measured annual average airborne plutonium concentrations resulting from resuspension. The basis is an annual average airborne mass loading of $260\,\mu g/m^3$ and a plutonium activity density on airborne soil corresponding to the upper limits for data shown in Fig. 1, i.e. activity densities from 4×10^3 to 1.3×10^5 fCi/g. Airborne plutonium concentrations are calculated by multiplying the mass loading by the activity density. Thus, the maximum estimated annual-average airborne plutonium concentrations from wind resuspension range from 1.0×10^{-15} to $3.4 \times 10^{-14}\,\mu$Ci/cm^3. To be emphasized is that these are conservative estimates directed toward illustrating the maximum values which might be predicted from the modified mass-loading calculational procedure. These estimates are for annual planning purposes, rather than for daily applications to site specific areas. For daily applications, the wind speed dependencies, etc. of resuspension physics must also be considered.

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Establishment of a Field Plot to Study the Behavior of Cesium-137 in the Environment

ALTHOUGH the behavior of ^{137}Cs in the environment has been the subject of many studies (De 78; Le75; Ro70; Au58; Al79), any new situation which can be followed closely may contribute to a better understanding. Such a situation presented itself during the summer of 1979 with an accidental release at an industrial site near Louisiana State University (LSU) when a 1–Ci ^{137}CsCl source ruptured during a recovery operation. Approximately one-third of the loose, powdered material was dissolved and washed onto adjoining grass sod and a pasture by a rainstorm before cleanup was completed.

As part of the decontamination efforts at the accident site, the Louisiana Nuclear Energy Division (LNED) ordered removal and disposal of the most severely contaminated surface material. The faculty at LSU's Nuclear Science Center was aware of the situation and recognized the opportunity to undertake field studies of ^{137}Cs movement in the environment. The center had an area previously used for burial of short-lived radionuclides, but which has been fallow for more than 20 yr, as a candidate site. Subsequently, LNED authorized transfer of approximately 35 m^2 of St. Augustine grass sod and 2 m^3 of underlying soil to LSU to establish a Radioecology Field Laboratory.

The site of the Radioecology Field Laboratory is a 166×10 m (0.5 ha) fenced area located on an ancient flood plain 1.5 km from the Mississippi River levee system on the LSU Agricultural Experiment Station. This fenced area has not been cultivated since 1954, and has been maintained only by occasional mowing.

The soil in this area is a low-permeability Mhoon

PLUTONIUM-239 AND AMERICIUM-241 CONTAMINATION IN THE DENVER AREA

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Abstract—Procedures and results of an experimental investigation of ^{239}Pu and ^{241}Am contamination in the environs of the Rocky Flats plutonium plant and elsewhere in the greater Denver area are presented and discussed. Measurements of ^{239}Pu and ^{90}Sr in the top 1 cm surface layer of soils show that in this layer the ^{239}Pu contamination in offsite areas just east of the Rocky Flats plant ranges up to hundreds of times that from nuclear tests. In the more densely populated areas of Denver the Pu contamination level in surface soils is several times fallout. The depth distribution of ^{239}Pu in undisturbed soils in the more heavily contaminated areas show that the contaminant is concentrated largely in a thin surface layer. Results for soils of known last date of disturbance make it evident that most of the offsite Pu accumulated between 1966 and 1969. This corresponds to the period in which Pu in an oil spill area on the plant site was exposed and subjected to wind reentrainment. ^{241}Am, which grows in from ^{241}Pu present in the contaminant, now contributes between 3 and 15% as much alpha activity as ^{239}Pu and ultimately will approach comparable levels.

The important unknowns and uncertainties involved in the estimation of inhalation exposures and the carcinogenic risks from the ^{239}Pu contamination in the environs of Rocky Flats are briefly reviewed.

1. INTRODUCTION

ON 11 May 1969 a fire involving a large amount of plutonium occurred at the Atomic Energy Commission's plutonium processing plant† at Rocky Flats, 16 miles northwest of downtown Denver. During the fire heavy black fumes were observed to discharge into the atmosphere for a brief period and a lighter grey-brown smoke issued from the building for a prolonged period of time. Subsequently millicurie quantities of ^{239}Pu were found on the roof of the building and on a paved area nearby. The amount of Pu released to the outside environment by this fire is not known.

When plutonium is burned in air the particles produced are highly insoluble PuO_2 particles of respirable size[1] and high specific alpha radioactivity. Inhaled particles of this type carry a carcinogenic risk of uncertain magnitude. The prescribed maximum permissible lung burden (MPLB) of 0.016 μCi ^{239}Pu for occupational exposure which is based on uniform tissue dose experience and a relative biological

effectiveness (RBE) factor of 10 for alpha radioactivity, may involve a substantial risk.[2,3]

At the outset of this experimental study, the objective was to determine the extent of offsite Pu contamination from the May 1969 fire. Our initial soil measurements, made during 1969, were reported elsewhere[4] and showed appreciable levels of ^{239}Pu in areas immediately east of the plant. Subsequently representatives of the Dow Chemical Company suggested a September 1957 Pu fire at the Rocky Flats plant[5] and a Pu contaminated oil spill on the plant site as other possible sources of the offsite Pu contamination.

Our ^{239}Pu measurements in soils, surface waters and sediments in offsite areas surrounding the plant are presented below. The soil samples also were subjected to ^{90}Sr assay to provide means of distinguishing between ^{239}Pu of Rocky Flats plant origin and ^{239}Pu fallout from nuclear tests. To assess which of the suggested sources was responsible for the offsite Pu, the depth distribution of ^{239}Pu and ^{90}Sr for a limited number of soils of known last date of disturbance was determined. ^{241}Am alpha radioactivity also is reported for some soil, water and sediment samples. ^{241}Am is the daughter product of

^{241}Pu, a β^- emitter of 13 yr half-life, present in the Rocky Flats plant plutonium isotope mixture.[6]

2. PROCEDURES

(a) Determination of ^{239}Pu (plus ^{240}Pu)* and ^{90}Sr

After preliminary processing of soil and sediment samples, 10 g fractions were taken for analysis. Both 60 mg of $Sr(NO_3)_2$ as carrier and a measured aliquot (~10 dpm) of ^{236}Pu calibrated standard solution were added. Each sample was dried and ignited to 600°C to remove volatiles. The sample was digested, in turn with 200 ml 6N HCl, 100 ml conc. HNO_3, 20 ml 30% H_2O_2 and successively larger amounts of water. The soil sample was filtered and washed twice with 2N HNO_3. To the combined filtrate and washes, 5 ml of concentrated H_3PO_4 was added. NaOH was slowly added until a precipitate formed and then the solution was adjusted to pH 9 with NH_4OH.[7,8] After prolonged digestion the mixture was centrifuged. The precipitate was redissolved in HNO_3 and the solution adjusted to 300 ml with 8N HNO_3. This solution was slowly passed (~100 ml/hr) through a freshly prepared Dowex 1-X2 (50–100 mesh) ion exchange column in the nitrate form. The column was washed with 8N HNO_3 and the HNO_3 solutions were combined and reserved for Sr analysis.[8]

The column was washed with 20 ml of concentrated HCl and the solution discarded. Pu was eluted with 250 ml 0.5N HCl and converted to the nitrate by repeated evaporations to dryness with added HNO_3.[7,9] The Pu was purified by recycling through the regenerated column when excess residue was observed.

(b) Determination of ^{241}Am and ^{239}Pu

Small aliquots of calibrated spike solutions of ^{236}Pu and ^{243}Am were added to 10 g of screened dry soil in a Pt dish and heated to 600°C. The sample was first digested in 6N HCl, then with conc. HNO_3, cooled and 10 ml 30% H_2O_2 was added. After the H_2O_2 reaction was complete, the sample was digested, cooled, diluted to 3N HNO_3 and centrifuged. The centrifugate was passed slowly (~100 ml/hr)

through a Dowex 1 column in the NO_3^- form; the column was washed with 8N HNO_3 and the HNO_3 solutions combined for Am separation. The Pu was removed from the column with 0.5N HCl and was converted to nitrate[7,9] and plated (see below). To the Am solution some La carrier was added. The solution was evaporated to dryness and converted to Cl^- with HCl. The residue was digested with 100 ml H_2O, centrifuged and the residue rejected. The solution acidity was reduced with NaOH and 20 ml saturated oxalic acid added. Solution was adjusted to pH ~2 with NaOH, digested, and oxalate precipitate was centrifuged, redissolved in 0.5N HCl, adsorbed on a Dowex 50-X8 (50–100 mesh) column and the Am eluted with HCl-ethanol solution.[10-12]

(c) Plutonium plating

The purified Pu nitrate solution was evaporated to dryness and 10 ml 0.4N HNO_3 was added. A polished stainless steel disk was washed with HNO_3, rinsed and placed in a plating cell. Two ml of 1.5N NH_4OH were added to the Pu solution and the solution poured into the plating cell. Pu was plated at the cathode over 0.8 cm² area of the stainless disk for a period of 3 hr at ~0.3 amp current and 12 V potential. The plating solution was made slightly basic during the last 30 min with NH_4OH added as necessary. The plated disk was subjected to red heat in an oxidizing flame. This procedure is essentially that of SANDERS and LEIDT.[13]

(d) Americium plating

The HCl-ethanol solution of Am was evaporated to dryness and the residue dissolved in 10 ml NH_4Cl plating solution.[14] The solution was adjusted to pH 3 with 2N HCl. The Am was plated on a Pt disk cathode at 12 V and 0.5 amp for 2 hr. Thick deposits which gave poor spectra were removed with concentrated HNO_3, recycled on Dowex 50 and/or Dowex 1 and replated.

(e) Strontium-90 determination

Following purification and yield determination, the Sr fraction was stored to allow ^{90}Y growth. Subsequently Y carrier was added and the Y was separated, purified,[8] and Y yield

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* Because their α energies are indistinguishable the abundant ^{239}Pu and less abundant ^{240}Pu are determined together but usually referred to as ^{239}Pu alone.

determined. The Y sample was mounted on a thin backing and counted between two thin-walled β^- flow counters in close sandwich geometry with steel and anticoincidence shielding. The ^{90}Y was counted in decay to allow resolution of the ^{90}Y activity from the chemical blank β^- activity and background. The absolute ^{90}Sr activity was calculated from the measured ^{90}Y activity and Y and Sr yield data, using techniques calibrated with a ^{90}Sr solution standard.

(f) Plutonium-239 and ^{241}Am measurement

The plated samples were mounted close to a silicon surface barrier crystal detector in a vacuum chamber at room temperature. Samples were counted with a multichannel pulse height analyzer. Calibrated standard spikes of ^{236}Pu and ^{243}Am which had been added to each samples for ^{239}Pu and ^{241}Am analysis respectively, eliminated uncertainties due to recovery yield, plating efficiency, adsorption, scattering and geometry. Absolute disintegration rates were estimated directly from the ratio of total pulses under the ^{239}Pu (plus ^{240}Pu) and ^{236}Pu peaks (or ^{241}Am and ^{243}Am peaks) and the known spike activity, with very small corrections for background and spike impurities.

(g) Soil sampling and processing

All soil samples (with the exception of soil profiles 3, 4, 5 and 7, Table 3) were collected from even, undisturbed sites which had not been subjected to the action of irrigation or plowing. Where the history of the sampling sites was not known the soil samples were taken along old fence lines near midspan between posts. The vegetation was cropped closely and discarded. Each soil sample was collected over an area about 1000 cm² by spatula, with special care to provide samples of the stated depth as uniformly as practical. For depth profiles precautions were taken to avoid contamination from the higher soil horizons.

The soil samples were air dried, pulverized to break down soil aggregates, sieved to removed particles >0.05 cm diameter and thoroughly mixed. For each analysis a 10 g aliquot was taken by the cone and quarter technique. The measured density of the screened and dried surface soils averaged about 1.0 g/cm³, the approximate density of the top 1 cm depth of soils. Thus a radioactivity concentration of 1 dpm/g in surface soils (Tables 1 and 2) corresponds to ~1 dpm/cm² (equivalent to 4.5 mCi/km²) in the 1 cm depth surface layer.

To test the adequacy of procedures for the extraction of ^{239}Pu and ^{90}Sr from soils, a number of samples were reextracted and yielded negligible additional ^{239}Pu and ^{90}Sr. Replicate analyses of a number of soil samples were made and all showed good agreement within twice the standard deviation except for samples at sites B and C, included in Table 2. The lack

Table 1. Plutonium-239 and ^{90}Sr fallout from nuclear tests in surface soils, Colorado Eastern Slope areas*

Location	^{239}Pu (dpm/g)	^{90}Sr (dpm/g)	^{239}Pu/^{90}Sr (activity ratio)
Loveland	0.047 ± 0.013	1.05 ● 0.02	0.044 ± 0.012
Loveland	0.056 ± 0.025	1.01 ± 0.03	0.055 ± 0.025
Loveland	0.045 ± 0.008	1.09 ± 0.02	0.041 ± 0.007
Loveland	0.026 ± 0.006	1.23 ± 0.02	0.021 ± 0.005
Loveland	0.043 ± 0.005	1.00 ± 0.01	0.043 ± 0.005
Brighton	0.093 ± 0.009	5.13 ± 0.04	0.018 ± 0.002
Cripple Creek	0.140 ± 0.027	3.01 ± 0.07	0.047 ± 0.009
Cripple Creek	0.052 ± 0.012	1.10 ± 0.02	0.047 ± 0.011
Cripple Creek	0.117 ± 0.015	3.76 ± 0.03	0.031 ± 0.004
		Weighted average†	0.034 ± 0.005

* Soil samples, 0–1.0 cm depth and ~1000 cm² area collected September 1969—January 1970 (Errors tabulated represent one standard deviation).

† Weighting is inversely proportional to the per cent error represented by one standard deviation.

Table 2. Plutonium-239 and ^{90}Sr in surface soils in the Denver area*

Location†	Collection date	^{239}Pu (dpm/g)	^{90}Sr (dpm/g)	^{239}Pu/^{90}Sr (activity ratio)
A	8/25/69	1.29 ± 0.06	1.13 ± 0.03	1.14 ± 0.06
B	8/25/69	2.33 ± 0.11	2.14 ± 0.01	1.08 ± 0.05
B	8/25/69	13.5 ± 0.9	1.59 ± 0.02	8.49 ± 0.58
C	8/25/69	0.092 ± 0.018	0.57 ± 0.02	0.16 ± 0.03
C	8/25/69	0.057 ± 0.014	0.72 ± 0.05	0.079 ± 0.020
D	9/18/69	0.128 ± 0.009	0.29 ± 0.01	0.44 ± 0.04
E	9/18/69	0.279 ± 0.015	4.08 ± 0.08	0.063 ± 0.004
F	9/18/69	0.209 ± 0.020	2.93 ± 0.07	0.0713 ± 0.007
G	10/2/69	0.314 ± 0.022	5.58 ± 0.10	0.0563 ± 0.004
H	10/2/69	0.129 ± 0.018	2.04 ± 0.03	0.063 ± 0.009
I	10/2/69	1.72 ± 0.08	0.45 ± 0.02	3.8 ± 0.2
J	10/2/69	1.15 ± 0.05	0.91 ± 0.04	1.3 ± 0.1
K	10/2/69	0.881 ± 0.044	1.75 ± 0.03	0.503 ± 0.027
L	10/2/69	0.116 ± 0.017	0.52 ± 0.01	0.22 ± 0.03
M	10/2/69	0.370 ± 0.030	1.31 ± 0.03	0.282 ± 0.024
N	10/2/69	1.34 ± 0.05	2.28 ± 0.08	0.588 ± 0.030
O	11/20/69	0.068 ± 0.011	1.96 ± 0.02	0.035 ± 0.006
P	11/20/69	0.108 ± 0.015	1.41 ± 0.02	0.077 ± 0.011
Q	11/20/69	0.040 ± 0.007	0.79 ± 0.02	0.051 ± 0.009
R	11/20/69	0.078 ± 0.016	1.00 ± 0.03	0.078 ± 0.016
S	11/20/69	0.102 ± 0.011	0.61 ± 0.02	0.17 ± 0.02
T	11/20/69	0.175 ± 0.038	—	—
U	11/20/69	0.320 ± 0.012	2.06 ± 0.02	0.16 ± 0.01
V	11/20/69	0.543 ± 0.027	1.80 ± 0.02	0.302 ± 0.015
W	11/20/69	0.033 ± 0.009	0.51 ± 0.02	0.065 ± 0.018
X	11/20/69	0.187 ± 0.017	2.38 ± 0.04	0.079 ± 0.007
Y	11/20/69	0.420 ± 0.025	3.53 ± 0.05	0.119 ± 0.007
Z_1	1/28/70	0.117 ± 0.022	2.79 ± 0.03	0.042 ± 0.008
Z_2	1/28/70	0.239 ± 0.041	1.54 ± 0.03	0.155 ± 0.027
Z_3	11/18/69	0.304 ± 0.023	4.84 ± 0.10	0.063 ± 0.005
Derby	1/28/70	0.052 ± 0.009	0.37 ± 0.01	0.14 ± 0.02
NE Denver	1/28/70	0.104 ± 0.013	1.07 ± 0.03	0.097 ± 0.013
Aurora	1/28/70	0.055 ± 0.016	0.34 ± 0.02	0.16 ± 0.05
E Denver	1/28/70	0.106 ± 0.018	0.58 ± 0.02	0.18 ± 0.03
SE Denver	1/28/70	0.091 ± 0.014	1.25 ± 0.02	0.073 ± 0.011
SW Denver	1/28/70	0.068 ± 0.009	1.15 ± 0.03	0.059 ± 0.008
Golden	1/28/70	0.088 ± 0.012	0.95 ± 0.02	0.093 ± 0.013

* Soil samples 0–1.0 cm depth and ~1000 cm² area (errors tabulated represent one standard deviation).
† Soil sampling sites near the Rocky Flats plant are indicated by capital letters on the map, Fig. 1.

of agreement of results for these two sites, which are near the contaminant source, are attributed to the presence of relatively large Pu particles. The difference in results for two 10 g samples from site B is equivalent to the alpha activity of one 6 μ diameter particle of ^{239}PuO$_2$.

3. RESULTS AND DISCUSSION

(a) *Plutonium-239 and ^{90}Sr in surface soils*

Our initial assessment of the presence of ^{239}Pu contamination in offsite areas surrounding the Rocky Flats plutonium plant involved the collection of surface soil samples of 1 cm depth. The samples were subjected to both ^{90}Sr and ^{239}Pu assay in order to provide basis for distinguishing between ^{239}Pu from the Rocky Flats plant and that from nuclear weapons tests. The ^{239}Pu/^{90}Sr activity ratio observed for airborne debris from nuclear tests ranges from 0.016 to 0.041.[15,16] The cumulative total of ^{239}Pu and ^{90}Sr fallout from nuclear tests deposited in Derby, Colorado up to 1970 is

reported[18,17] to be 1.5 mCi [239]Pu/km[2] and 65 mCi ^{90}Sr/km[2]. This corresponds to a [239]Pu/^{90}Sr activity ratio of 0.023 for the cumulative fallout (averaged over its full depth distribution) in the Denver area soils. However because Derby is only 17 miles east of Rocky Flats and appears to have received some [239]Pu from the plant (Table 2 and discussion), 0.023 perhaps should be considered an upper limit for the cumulative fallout ratio. HARLEY[19] reported a [239]Pu/^{90}Sr production ratio of 0.017 for Northern Hemisphere fallout, corresponding to a ratio of 0.023 for cumulative fallout in 1971 after correction for ^{90}Sr decay. The [239]Pu/^{90}Sr activity ratios for the top 1 cm depth layer of soils in Colorado eastern slope areas at some distance from Rocky Flats, Table 1, range from 0.018 to 0.055 with a weighted average of 0.034 ± 0.005. This average ratio of 0.034 for surface soils is higher than the ratio 0.023 for cumulative fallout. The difference may be explained either by the more rapid transport of water soluble ^{90}Sr to greater depths by the action of soil moisture than is the case for insoluble [239]Pu or by the presence of [239]Pu from sources of different [239]Pu/^{90}Sr ratio.

Results for surface soils collected in offsite areas around the plant at locations shown in Fig. 1 and elsewhere in the Denver area are listed in Table 2. The highest [239]Pu concentrations were observed in areas east of the plant (Table 2 and Fig. 1).

The contribution of [239]Pu from Rocky Flats relative to that from nuclear tests can be estimated by dividing the [239]Pu/^{90}Sr ratios (last column, Table 2) by 0.034, the average [239]Pu/^{90}Sr ratio for surface soils at more distant locations (Table 1), and subtracting 1. Thus the Rocky Flats contribution ranges from 250 times fallout at a distance of 2 miles east of the plant (site B) to 10 times fallout in Westminster, 8 miles east of the plant (site U) and to several times fallout in the eastern suburbs of Denver (Table 2). These estimates apply only for the [239]Pu present in the top 1 cm depth of undisturbed soils.

(b) Plutonium-239 versus depth

The depth distribution of [239]Pu and ^{90}Sr was determined in soil samples collected from se-lected sites in the contaminated area east of the Rocky Flats plant and the results are presented in Table 3. Profiles number 1, 2 and 6, Table 3, were collected in undisturbed grassy areas and show that most of the [239]Pu contamination from Rocky Flats is concentrated in a thin layer at the soil surface. On first consideration the data for these three undisturbed profiles suggest that ^{90}Sr is transported downward more rapidly than [239]Pu by the action of soil moisture and other weathering processes. However even the lowest soil horizons show [239]Pu/^{90}Sr ratios (Table 3, last column) which equal or exceed 0.023, the [239]Pu/^{90}Sr ratio for cumulative fallout from nuclear tests. Thus we have no unequivocal evidence that ^{90}Sr is transported downward more rapidly than [239]Pu in undisturbed soils in Colorado eastern slope areas. The concentration of [239]Pu at the soil surface in areas immediately east of Rocky Flats is attributed to the recent arrival of [239]Pu contamination from the plant site (see below).

KREY and HARDY[17] report a large fraction of the total [239]Pu at greater soil depths. However this observation applies only for soil samples from locations some distance from the more heavily contaminated area east of the plant or for disturbed soils.

Soil samples for profile number 3, Table 3, were collected on 20 November 1969 at a fence line in a roadside depression at site I. For this profile, the low surface concentration of [239]Pu compared to that of a surface sample collected at this site on 2 October 1969 (Table 2) as well as the irregular depth distribution of the [239]Pu were, at first, difficult to explain. When this site was revisited again in January 1970, it was observed that more than 3 ft of dirt had accumulated in some sections of this roadside depression as a result of wind erosion of a large plowed area west of the site. It is evident that a depression such as site I is an area which accumulates windblown soil material and thus is a disturbed site, unsuitable for soil sampling.

(c) Offsite contaminant origin

In an attempt to date the period of arrival of the offsite contamination and thereby establish the source of the offsite [239]Pu, two soil profiles were collected on an earth dam between sites J and L, Fig. 1, and two others nearby
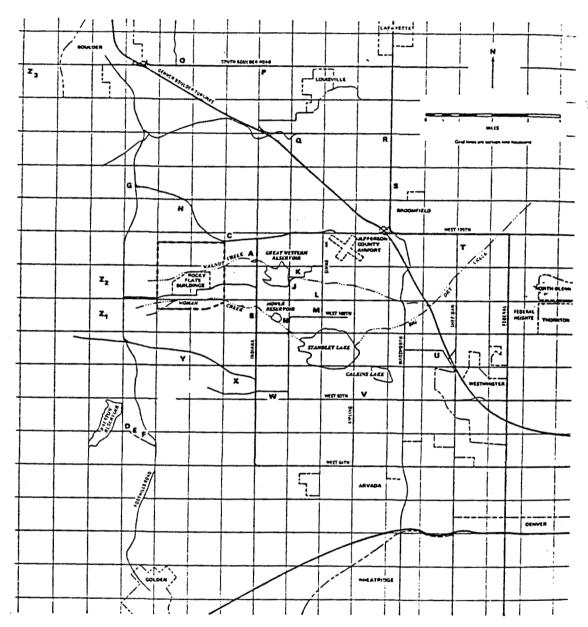
FIG. 1. Dow Chemical Company Rocky Flats plutonium plant and surrounding areas. Capital
letters indicate soil sampling sites (see Tables 2 and 3 for ^{239}Pu and ^{90}Sr results).

(profiles 4–7, Table 3). The accumulation of ^{239}Pu on the earth dam (profiles 4 and 5, Table 3) amounts to only a few per cent of the total ^{239}Pu accumulated at nearby sites (profiles 6 and 7, Table 3). It is evident that most of the offsite contamination east of the Rocky Flats plant was deposited before April 1969 when construction of the earth dam was completed. Thus the 11 May 1969 fire at the Rocky Flats plant could not have contributed significantly to the offsite ^{239}Pu contamination in this area. The ^{239}Pu on the earth dam provides an indication of the amount accumulated due to local redistribution of surface soil contamination in

Table 3. Vertical profiles of ^{239}Pu and ^{90}Sr in soils in the contaminated area east of Rocky Flats

Profile No.	Location* (date collected)	Depth (cm)	^{239}Pu (dpm/g)	^{90}Sr (dpm/g)	^{239}Pu/^{90}Sr (activity ratio)
1	J	0–0.3	4.64 ± 0.40	2.61 ± 0.07	1.78 ± 0.16
	(12/17/69)	0.3–1.3	1.73 ± 0.09	2.67 ± 0.06	0.648 ± 0.037
		1.3–2.5	0.070 ± 0.011	1.72 ± 0.04	0.041 ± 0.007
		2.5–5.0	0.018 ± 0.005	1.26 ± 0.03	0.014 ± 0.004
		5.0–7.5	0.020 ± 0.006	0.39 ± 0.02	0.051 ± 0.016
		7.5–10.0	0.017 ± 0.008	0.13 ± 0.01	0.13 ± 0.06
2	K	0–0.3	5.02 ± 0.17	1.62 ± 0.03	3.10 ± 0.12
	(12/17/69)	0.3–1.3	0.89 ± 0.13	1.28 ± 0.04	0.70 ± 0.10
		1.3–2.5	0.121 ± 0.030	1.31 ± 0.01	0.092 ± 0.023
3	I	0–0.3	0.090 ± 0.022	0.77 ± 0.01	0.12 ± 0.03
	(11/20/69)	0.3–0.7	0.049 ± 0.010	0.60 ± 0.02	0.082 ± 0.017
		1.3–2.5	0.044 ± 0.006	0.56 ± 0.02	0.078 ± 0.011
		5.0–7.5	0.078 ± 0.019	0.70 ± 0.03	0.11 ± 0.03
		12.7–14.0	0.096 ± 0.016	0.58 ± 0.01	0.17 ± 0.03
4	Site No. 1 on Earth Dam†	0–0.3	0.050 ± 0.013	0.15 ± 0.01	0.33 ± 0.09
	between J and L con-	0.3–1.3	0.047 ± 0.018	0.23 ± 0.02	0.20 ± 0.08
	structed in March and April	1.3–2.5	0.046 ± 0.005	0.16 ± 0.02	0.29 ± 0.05
	1969	2.5–5.0	0.006 ± 0.005		
	(2/10/70)	5.0–7.5	0.009 ± 0.005		
5	Site No. 2 on Earth Dam†	0–0.3	0.182 ± 0.008	0.33 ± 0.02	0.56 ± 0.04
	(2/10/70)	0.3–1.3	0.029 ± 0.007	0.18 ± 0.01	0.16 ± 0.04
		1.3–2.5	0.057 ± 0.018	0.08 ± 0.01	0.7 ± 0.2
		2.5–5.0	0.014 ± 0.004	0.06 ± 0.01	0.2 ± 0.1
		5.0–7.5	0.068 ± 0.014	0.13 ± 0.02	0.52 ± 0.13
		7.5–10.0	0.010 ± 0.005	0.02 ± 0.02	0.5 ± 0.6
6	Area 600′ from Earth Dam	0–0.3	2.39 ± 0.10	2.08 ± 0.03	1.15 ± 0.05
	undisturbed at least 8 yr	0.3–1.3	0.512 ± 0.036	1.67 ± 0.04	0.307 ± 0.023
	(2/10/70)	1.3–2.5	0.048 ± 0.006	1.13 ± 0.03	0.043 ± 0.005
		2.5–5.0	0.019 ± 0.005	0.75 ± 0.08	0.025 ± 0.007
		7.5–10.0	0.004 ± 0.004	0.34 ± 0.02	0.01 ± 0.01
7	Irrigated pasture area 300′	0–0.3	1.62 ± 0.08	0.78 ± 0.04	2.1 ± 0.2
	from Earth Dam last	0.3–1.3	0.82 ± 0.04	0.52 ± 0.02	1.6 ± 0.1
	ploughed in 1966 or 1967	1.3–2.5	2.07 ± 0.08	0.39 ± 0.01	5.3 ± 0.2
	(2/10/70)	2.5–5.0	0.159 ± 0.027	0.71 ± 0.02	0.22 ± 0.04
		5.0–7.5	0.060 ± 0.015	0.57 ± 0.03	0.11 ± 0.03
		7.5–10.0	0.107 ± 0.007	0.54 ± 0.03	0.20 ± 0.02

* Locations indicated by capital letters are shown on the map, Fig. 1.
† Surface samples of 0.3 cm depth from 3 other sites on the Earth Dam showed 0.076 ± 0.013, 0.133 ± 0.016 and 0.080 ± 0.008 dpm ^{239}Pu/g soil.

the 10 month period between dam construction and sample collection. However because of the unusual configuration of the terrain at the dam site and the presence of exposed subsoil immediately upwind, this apparent rate of redistribution may not represent that for more regular and undisturbed terrain.

The very irregular depth distribution of ^{239}Pu and ^{90}Sr in soil profile 7, Table 3, compared to their smooth trend in undisturbed soil profiles 1 and 6, Table 3, may be due to the action of irrigation as well as plowing. The high ^{239}Pu in this profile is confined to the upper 2.5 cm depth, indicating that most of the offsite Pu contamination had accumulated at this location since it was plowed in 1966. Thus the 1957 fire did not contribute substantially to the more heavily contaminated area east of the plant.

The period of accumulation of most of the offsite [239]Pu contamination east of the plant, 1966 to April 1969, corresponds closely to the period the oil spill area contamination was exposed to the action of winds, between 1967 when the drums were reprocessed and July 1969 when the spill area was paved.[20] Thus it is concluded that Pu from the spill area was the main source of contamination in the contoured area, Fig. 2.

(d) *Plutonium-239 in surface waters and sediments*

Results of [239]Pu measurements in local surface waters and sediments are summarized in Table 4. The highest levels of [239]Pu are observed in the pond at A (Fig. 1) and in Great Western Reservoir, part of the Broomfield water supply. Both are fed by Walnut Creek which receives liquid waste effluents from the Rocky Flats plant after they have passed through several liquid waste holding ponds on the plant site. The results show that [239]Pu is highly adsorbed on sediments and suggest that the [239]Pu in liquid effluents may be largely concentrated on sediments within the holding ponds and along Walnut Creek and perhaps in the pond at A as well. Levels of [239]Pu in mud and silt of Great Western Reservoir are comparable to [239]Pu in nearby surface soils (Table 2). Thus

[239]Pu already present in Great Western Reservoir may be explained by airborne debris from the spill area rather than by accumulation of liquid wastes.

Local surface waters all show [239]Pu concentrations appreciably higher than that of Boyd Lake in Loveland, Colorado. However, results for Boyd Lake, about 40 miles north of Rocky Flats, are not necessarily representative of [239]Pu levels from nuclear tests, The maximum permissible concentration of [239]Pu in water applicable to exposure of the general public is $5 \times 10^{-6} \mu Ci/ml$,[21] corresponding to 11×10^3 dpm/l. Thus the observed range of concentrations of [239]Pu in surface waters, Table 4, are well below the recommended standard. However because [239]Pu is concentrated by adsorption on sediments, uptake of [239]Pu in fish, plants, and other biological material merits investigation. Algae taken from Great Western Reservoir contained 2.1 dpm [239]Pu per gram of dried algae (Table 5), a level appreciably higher than that of sediment samples from this reservoir (Table 4).

(e) *Americium-241 contamination*

A full appreciation of the nature and significance of the alpha radioactivity contamination

Table 4. Plutonium-239 in surface waters and sediments

Sample location	Collection date	[239]Pu in water (dpm/l.)†	[239]Pu in sediments (dpm/g)†
Pond A*	8/25/69	0.432 ± 0.030	6.99 ± 0.30
Pond A*	8/25/69	—	18.2 ± 1.2
Pond A*	8/25/69	—	4.82 ± 0.21
Pond A*	7/15/70	1.81 ± 0.18	—
Great Western Reservoir*	10/2/69	0.214 ± 0.007	0.142 ± 0.016
Great Western Reservoir*	7/15/70	0.046 ± 0.008	0.454 ± 0.051
Calkins Lake*	9/10/69	0.016 ± 0.004	0.109 ± 0.009
Ralston Reservoir*	9/18/69	0.025 ± 0.004	—
Boulder Reservoir (NE of Boulder)	9/11/69	0.016 ± 0.016	0.015 ± 0.004
Dodd's Lake (NE of Boulder)	8/31/69	0.023 ± 0.004	—
	9/11/69	0.029 ± 0.014	—
Boyd Lake (Loveland, Colo.)	9/22/69	0.003 ± 0.002	0.014 ± 0.004
Tap water, Broomfield, Colo.‡	2/5/70	0.038 ± 0.005	—

* Locations shown on map, Fig. 1.
† Errors tabulated represent one standard deviation.
‡ Broomfield draws part of its water supply from Great Western Reservoir.

PLUTONIUM-239 CONTOURS, mCi/km²

FIG. 2. Plutonium-239 accumulation in soils surrounding the Dow Chemical Company Rocky Flats plutonium plant, KREY and HARDY.[17]

544

Table 5. *Americium-241 and* [239]Pu *in environmental samples*

Sample description*	Collection date	[241]Am (dpm/g)†	[239]Pu (dpm/g)†	[241]Am/[239]Pu (activity ratio)
Soil, E	9/18/69	0.030 ± 0.013	0.246 ± 0.021	0.12 ± 0.07
Soil, J	10/2/69	0.30 ± 0.04	7.75 ± 0.95	0.04 ± 0.01
Soil, N	10/2/69	0.13 ± 0.02	0.90 ± 0.05	0.14 ± 0.02
Soil, U	11/20/69	0.030 ± 0.013	0.89 ± 0.05	0.034 ± 0.015
Soil, (surface Profile 6, Table 3)	2/10/70	0.48 ± 0.07	2.88 ± 0.34	0.17 ± 0.03
Sediment (Pond at A)	8/25/69	2.8 ± 0.3	18.2 ± 1.2	0.15 ± 0.05
Sediment (Great Western)	10/2/69	0.06 ± 0.03	0.19 ± 0.03	0.3 ± 0.2
Sediment (Great Western)	10/2/70	0.07 ± 0.04	0.45 ± 0.05	0.2 ± 0.1
Algae, genus *Rhizoclonium* (Great Western)	10/2/70	0.26 ± 0.07	2.12 ± 0.21	0.12 ± 0.04
Water (Pond at A)	7/15/70	0.43 ± 0.13‡	1.81 ± 0.18‡	0.24 ± 0.08

* Locations shown in Fig. 1.
† Errors tabulated represent one standard deviation.
‡ Activity in dpm/l.

west of Denver requires the assessment of present and future levels of [241]Am and its distribution in the environment. Americium-241 is an alpha emitting isotope of the man-made transuranium element, americium. The presence of [241]Am in the Denver area contamination stems from its growth from [241]Pu, present in the Rocky Flats plutonium mixture. Owen[6] reports that most of the plutonium processed at Rocky Flats has an isotopic composition as follows: <1% [238]Pu, ~93% [239]Pu, ~6% [240]Pu, <1.0% [241]Pu and <0.1% [242]Pu. The [241]Pu decays by β^- emission with a half-life of 13 yr. The [241]Am, which emits 5.5 MeV α particles with a half-life of 460 yr, will attain its maximum radioactivity contribution when the [241]Pu is substantially depleted by radioactive decay, about 70–80 yr after chemical purification of the plutonium mixture. At that time the [241]Am alpha activity which would correspond to an initial [241]Pu abundance of 1.0% would add another 54% to the total α activity of Pu. For lower [241]Pu abundances the [241]Am would be correspondingly lower. However these estimates do not include the contribution of independent releases of [241]Am in liquid and airborne effluent which may result from the processing of chemically separated [241]Am at the Rocky Flats plant.[6]

Our results for [241]Am and [239]Pu measurements of environmental samples from the contaminated area east of Rocky Flats are summarized in Table 5. Our separation procedure for Am gave low recovery yields, which, coupled with the low total sample activities, resulted in large errors for some of the [241]Am results. The results (Table 5) show that present levels of [241]Am range between 3 and 30% of the [239]Pu activity. The variable concentration ratios are not inconsistent with [241]Am growth in Pu contaminant mixtures of various ages (times since Pu purification within the 20 yr period of Rocky Flats plant operation) and with various [241]Pu parent abundances, less than 1% of the Pu isotope mixture. It also is possible that differences in the chemical properties of [241]Am and [239]Pu give rise to their fractionation or separation by action of physical, chemical, and biological processes in the environment. Thus as the offsite contamination continues to age and the [241]Am increases due to [241]Pu decay, the [241]Am activity may approach or even exceed the [239]Pu activity of some environmental materials. For this reason it is important to carry out [241]Am as well as [239]Pu measurements in environmental surveys in the future in order to assess the full extent of [241]Am growth as well as to evaluate weathering and fractionation processes.

(*f*) *Plutonium-239 distribution*

Results of an independent study of the [239]Pu and ^{90}Sr in soils surrounding the Rocky Flats plant, reported by Krey and Hardy[17,18] provide added information on the contaminant

distribution. Krey and Hardy collected soil samples to a depth of 20 cm and thus their results should represent the total cumulative deposition, or nearly so. Their results for the ^{239}Pu distribution in the more heavily contaminated areas east of the plant are summarized in the contour map, Fig. 2. Based on soil measurements at Derby, Colorado, fallout from nuclear tests has contributed about 1.5 mCi ^{239}Pu/km^2 in this area. Thus the Rocky Flats plant contribution to the total ^{239}Pu contamination, Fig. 2, ranges from about 1300 times fallout at the east edge of the plant (the 2000 mCi/km^2 contour) to about equal that of fallout at the 3 mCi/km^2 contour. Our depth distribution of ^{239}Pu for undisturbed soils (profiles 1, 2, 6, Table 3) show that ^{239}Pu contamination from the plant is largely confined to the upper 1 or 2 cm depth within the contoured area, Fig. 2. When the depth distribution of the ^{239}Pu and the location of each soil sample is considered, our results, Table 2, and those of KREY and HARDY,[17,18] Fig. 2, show reasonably good agreement.

KREY and HARDY[17] estimate that the total ^{239}Pu from Rocky Flats plant in offsite areas within the 3 mCi/km^2 contour, Fig. 2, amounts to 2.6 Ci. Based on the ^{239}Pu and ^{90}Sr soil data[17,18] and taking 0.023 as the ^{239}Pu/^{90}Sr activity ratio for cumulative fallout, we estimate an additional 4.0 Ci of ^{239}Pu from Rocky Flats beyond the 3 mCi/km^2 contour out to 64 km distance in the northeast quadrant alone. On the same basis, the HASL results[17,18] for soils from Loveland, Plattville and LaSalle, Colorado (sites 48–65 km north and northeast of Rocky Flats) exhibit an average of 1.25 mCi ^{239}Pu/km^2 attributable to Rocky Flats. The latter result indicates an appreciable contribution of ^{239}Pu from the plant at and beyond 64 km distance to the northeast.

There are too few soil data to allow estimation of the amounts of ^{239}Pu contamination from the plant outside the 3 mCi/km^2 contour in the southeast, southwest and northwest directions. Winds during the 11 September 1957 and 11 May 1969 fires would have carried the effluent Pu to the southeast and southwest, respectively. Thus the assessment of the total offsite ^{239}Pu contamination is very incomplete.

4. CONCLUDING REMARKS

On the basis of the results presented in this paper it is concluded that most of the ^{239}Pu contamination in the contaminated offsite area east of the Rocky Flats plant was deposited in the period 1966–1969, indicating that the ^{239}Pu contaminated oil spill area on the plant site must be the main source. It also is clear that in undisturbed soils within the contaminated area most of the several curies of ^{239}Pu present is concentrated in a shallow layer at the soil surface. The offsite ^{239}Pu resulted from the wind re-entrainment of ^{239}Pu on the ground surface in the spill area on the plant site. The ^{239}Pu on soil surfaces in both onsite and offsite areas is subject to further redistribution by the action of winds. The possible significance of the offsite ^{239}Pu contamination stems mainly from the inhalation exposures which occurred during its initial transport to offsite areas plus the added exposures resulting from subsequent resuspension of the deposited ^{239}Pu by the further action of winds as well as by vehicular traffic and other activity. To the radiation dose from inhaled ^{239}Pu must be added that contributed by ^{241}Am, ultimately an additional 25–50%.

That inhalation exposures incurred during the period in which the offsite ^{239}Pu contamination was accumulating may have been significant is indicated by approximate dose commitment estimates. SHLEIEN[22] estimated a 50 yr dose commitment of 160 mrem to the respiratory lymph nodes of people due to fallout ^{239}Pu inhalation between May 1965 and March 1969. Over the full period of over 20 yr for all nuclear testing the corresponding 50 yr dose commitment would be about 10 times greater and therefore in excess of 1 rem. Within the contoured area, Fig. 2, where the ^{239}Pu contaminant from Rocky Flats ranges from 2 to hundreds of times the total fallout ^{239}Pu accumulation, the dose commitment would be correspondingly higher. Such an estimate assumes that the same proportion of ^{239}Pu particles from Rocky Flats and from fallout are of respirable size. Although particles released by Pu fires are known to be of respirable size,[1] the size distribution of the airborne ^{239}Pu transported from the spill area is not known.

The added inhalation exposures and dose commitments resulting from the disturbance of

contamination on soil surfaces are difficult to assess. That wind reentrainment of surface deposits takes place in the Rocky Flats area is amply illustrated by the fact that ^{239}Pu deposited on the ground in the oil spill area on the plant site accounts for most of the offsite contamination east of the plant. The physical processes involved in the resuspension of deposited particles are described by BAGNOLD.[23] The erosion of soil surfaces and the resuspension of Pu particles can proceed effectively by the action of strong, gusty winds on dry, sparsely vegetated surfaces—conditions which are commonplace at Rocky Flats and its environs. Because most of the offsite Pu contaminant east of the Rocky Flats is concentrated at the soil surface it can be subjected to the wind reentrainment process repeatedly until it is extensively weathered or plowed. Contaminant deposited on paved surfaces and elsewhere in urban areas is more readily disturbed and resuspended by vehicular traffic and human activity. An understanding of these processes and of the inhalation exposures resulting from them is needed to assess the significance of ^{239}Pu deposited on soils and other surfaces.

The distribution and origin of ^{239}Pu in soils at greater distances from the Rocky Flats plant merits further investigation. Because effluent from the 1957 aad 1969 fires would have been carried to the southeast and the southwest respectively, the distribution of ^{239}Pu in soils out to greater distances in these two quadrants is needed to assess the total offsite ^{239}Pu and the possible presence of areas of appreciable ^{239}Pu contamination. In addition, the presence of ~1.25 mCi ^{239}Pu/km^2 attributable to Rocky Flats at distances 48–65 miles north and northeast of the plant require further investigation and explanation. Plutonium-239 from the oil spill area would be disturbed only by strong, gusty winds and carried eastward, conforming to the pattern of offsite contamination, Fig. 2. If the presence of curie quantities of ^{239}Pu at appreciable distances to the north and northeast were to be explained by the accumulation of normal stack effluent from the plant, then the total of such effluents would exceed the reported total of 0.038 Ci[24] by more than two orders of magnitude.

No reliable estimate can be made of the ^{239}Pu lung burdens acquired during the period of offsite ^{239}Pu accumulation or due to its subsequent reentrainment In addition, even if the lung burdens were known, the cancer risk applicable to a given number and size of ^{239}Pu particles in the lung and lymph nodes is highly uncertain. For discussion of this complex problem (which includes the possibility that the present permissible lung burden, 0.016 μCi of ^{239}Pu, involves a high risk) the reader is referred to several recent articles.[2,3,25,26] In view of the possibly significant inhalation exposures to people working in and downwind of the spill area or living in areas east of the plant, a medical follow-up of the exposed groups, to assess Pu lung burdens and their consequences, would appear to be in order. Occupational groups with potentially high exposures include those who worked in the oil spill area, handling and reprocessing oil drums and those who were involved in paving the spill area. We recommend that the ^{239}Pu lung and lymph node burdens of domestic animals that have lived within the 10 mCi/km^2 contour area, Fig. 2, since 1966 be measured. Results of such measurements would provide a qualitative index for the inhalation exposures of the residents of this area.

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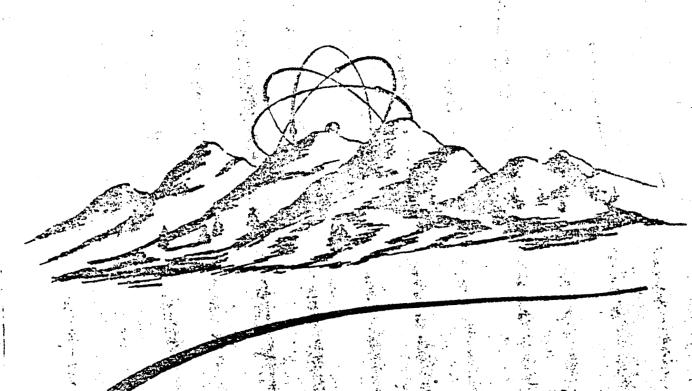
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DATA USED IN HEALTH PHYSICS CONSIDERATIONS
FOR PLUTONIUM AND AMERICIUM

by

E. A. Putzier

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NUCLEAR CONSTANTS

Isotope	Radiation	Energy	Abundance
Pu^{238}	α	5.495 Mev	72%
	α	5.452 Mev	28%
	α	5.352 Mev	0.09%
	L x-ray	17 kev avg	0.13/α decay
	γ-ray	45 kev	3.4 × 10^{-4}/α decay

Radiologic half-life   89.6 yr
Spontaneous fission half-life   3.8 × 10^{10} yr
Specific activity   16.8 c/g
Specific spontaneous fission rate   1140 f/g/sec
First daughter product — U^{234} (not important to health physics considerations)

Some small amount of hard γ associated with fission.

Isotope	Radiation	Energy	Abundance
Pu^{239}	α	5.147 Mev	72.5%
	α	5.134 Mev	16.8%
	α	5.096 Mev	10.7%
	L x-ray	17 kev avg	2.9 × 10^{-2}/α decay
	γ-ray	39 kev	2 × 10^{-5}/α decay
	γ-ray	53 kev	7 × 10^{-6}/α decay
	γ-ray	100 kev	5.5 × 10^{-6}/α decay
	γ-ray	124 kev	2.5 × 10^{-6}/α decay
	γ-ray	384 kev	1.5 × 10^{-5}/α decay

Radiologic half-life   24,400 yr
Spontaneous fission half-life   5.5 × 10^{15} yr
Specific activity   0.062 c/g
Specific spontaneous fission rate   0.02 f/g/sec
First daughter product — U^{235} (not important to health physics considerations)

Isotope	Radiation	Energy	Abundance
Pu^{240}	α	5.162 Mev	76%
	α	5.118 Mev	24%
	L x-ray	17 kev avg	0.1/α decay
	γ-ray	44 kev	10^{-4}/α decay

Radiologic half-life   6600 yr
Spontaneous fission half-life 1.2×10^{11} yr
Specific activity   0.227 c/g
Specific spontaneous fission rate   460 f/g/sec
First daughter product — U^{236} (not important to health physics considerations)

Some small amount of hard γ associated with fission.

Isotope	Radiation	Energy	Abundance
Pu^{241}	β^-	21 kev max	99+%
	α	4.9 Mev	4×10^{-3}%
	γ-ray	145 kev	2×10^{-4}/α decay

Radiologic half-life   13.2 yr
Specific activity   113 c/g
First daughter products — Am^{241} and U^{237} (discussed separately)

Isotope	Radiation	Energy	Abundance
Pu^{242}	α	4.898 Mev	76%
	α	4.858 Mev	24%

Radiologic half-life 3.8×10^5 yr
Spontaneous fission half-life 7×10^{10} yr
Specific activity 3.91×10^{-3} c/g
Specific spontaneous fission rate   2760 f/g/sec
First daughter product — U^{238} (not important to health physics considerations)

Isotope	Radiation	Energy	Abundance
Am²⁴¹	α	5.477 Mev	85%
	α	5.435 Mev	12.6%
	α	5.378 Mev	1.7%
	α others		<1%
	L x-ray	17 kev avg	3.7×10^{-1}/α decay
	γ-ray	26 kev	2.7×10^{-2}/α decay
	γ-ray	43 kev	6×10^{-4}/α decay
	γ-ray	60 kev	3.7×10^{-1}/α decay
	γ-ray	90 kev	2×10^{-4}/α decay

Radiologic half-life   462 yr
Specific activity   3.22 c/g
First daughter product — Np²³⁷ (not important to health physics considerations
of Am²⁴¹ chain)

Isotope	Radiation	Energy	Abundance
U²³⁷	β⁻	250 kev max	74%
	β⁻	84 kev max	26%
	γ-ray	59 kev	3.6×10^{-1}/α decay
	γ-ray	65 kev	2.3×10^{-2}/α decay
	γ-ray	165 kev	3.6×10^{-2}/α decay
	γ-ray	208 kev	2.4×10^{-1}/α decay
	γ-ray	268 kev	8.6×10^{-3}/α decay
	γ-ray	332 kev	1.6×10^{-2}/α decay

Radiologic half-life   6.7 days
Specific activity 8.2×10^4 c/g
First daughter product — Np²³⁷ (not important for health physics considerations
of Pu²⁴¹ chain)

CHANGING ACTIVITY IN A MIXTURE OF Pu ISOTOPES

The decay of Pu²⁴¹ to Am²⁴¹ and U²³⁷ has a decided influence on the over-all x- and γ-activity of a mixture of plutonium isotopes and to a lesser extent an influence on the α specific activity of the mixture. This influence can be predicted if the plutonium isotopic analysis of the mixture is known and the existing amount of U²³⁷ and Am²⁴¹ is known. The actual abundances of x- and γ-rays for a mixture consisting of 94.5% Pu²³⁹, 5% Pu²⁴⁰ and 0.5% Pu²⁴¹ with respect to time are shown in Figures 1 and 2. Values for other isotopic ratios would be in direct proportion to the isotopic ratios cited on these graphs. Figure 3 shows the changing activity of the same mixture with time. Again, values for other isotopic ratios would be in direct proportion to the isotopic ratios exhibited on these graphs. The very small α activity contribution from Pu²⁴¹ is ignored.

GAMMA DOSE RATES FROM "SEMI-INFINITE" THICK PURE METAL SLABS

1. Pu²³⁸

   L x-ray surface dose rate   980 Rad/

   45-kev γ surface dose rate   2 Rad/

2. Pu²³⁹

   L x-ray surface dose rate   0.61 Rad/

   γ radiations surface dose rate   0.056 Rad/

3. Pu²⁴⁰

   L x-ray surface dose rate   14 Rad/

44-kev γ-ray surface dose rate   8 mrad/hr

Hard γ-ray associated with fission
— surface dose rate   0.062 Rad/hr

4. Pu^{241} + daughters (Am^{241} and U^{237})

Surface dose rate from L x-rays from Am^{241} growth:

t < 2 years

$$230 (4.64 \times 10^{-4} t) \text{ Rad/hr} \qquad \text{t in days}$$

For all t

$$230 [3.33 (e^{-1.5 \times 10^{-3} t} - e^{-5.24 \times 10^{-2} t})] \text{ Rad/hr}$$

t in years

Surface dose rate from γ-rays from Am^{241} growth:

t < 2 years

$$221 (4.64 \times 10^{-4} t) \text{ Rad/hr} \qquad \text{t in days}$$

For all t

$$221 [3.33 (e^{-1.5 \times 10^{-3} t} - e^{-5.24 \times 10^{-2} t})] \text{ Rad/hr}$$

t in years

Surface dose rate from γ-rays from U^{237} growth:

For t < 40 days

$$24 (1 - e^{-0.102 t}) \text{ Rad/hr} \qquad \text{t in days}$$

For t > 40 days

$$24 \, e^{-\frac{0.693 t}{13.2 \times 365}} \text{ Rad/hr} \qquad \text{t in days}$$

Each of the above radiation levels is from a slab of metal of a single, initially pure isotope of Pu. Therefore, the values for a mixture must be weighted according to the isotopic make-up of the mixture. Figures 4 through 10 may be used for making a quick estimate of the surface dose rate from a slab of plutonium metal if the isotopic make-up and age since purification are known. Values for the contributions to the surface dose rate from Pu^{238}, Pu^{239} and Pu^{240} may be taken directly from Figures 4, 5, and 6, respectively. To obtain the contribution from the Am^{241} daughter of Pu^{241}, obtain the surface dose rate factor from Figure 8 and multiply by the appropriate time factor from Figure 7 (time < 2 years). To obtain the contribution from the U^{237} daughter

of Pu^{241}, obtain the surface dose rate factor from Figure 10 and multiply by the appropriate time factor from Figure 9 (time < 35 days, however, from the practical standpoint if 35 < t < 1 year, the value from Figure 10 may be used without a time factor; for t > 1 year, this value should be multiplied by

$$e^{-\frac{0.693 t}{13.2}}$$

with t in years). The five values may be added to estimate the gamma surface dose rate from the mixture.

NEUTRON RADIATION FROM Pu

The neutron radiation value (ngs) from pure plutonium metal depends primarily on the spontaneous fission rates of the isotopes which make up the mixture. Specific neutron yields are as follows:

Pu^{238} 3420 neutrons/g/sec
Pu^{239} Negligible
Pu^{240} 1380 neutrons/g/sec
Pu^{242} 2580 neutrons/g/sec

The ngs value for the plutonium mixtures common to Rocky Flats operations is, for practical purposes, dependent on the Pu^{240} content only. Figure 11 is a plot of ngs versus percent Pu^{240} in the mixture.

The neutron dose rate at the surface of a Pu sphere can be shown to be $\frac{1}{2}na$ where a is the radius of the sphere and n is the number of neutrons emitted per unit volume. For fission neutrons, $1 \text{ n/cm}^2/\text{sec} = 0.115 \text{ mrem/hr}$.

Neutrons may also be produced by the α-n nuclear reaction when Pu and certain light nuclei are combined as mixtures or compounds. Observed neutron yields from polonium alpha particles bombarding thick targets are shown in Table I. Yields from plutonium alpha particles may vary slightly from these, e.g., Rocky Flats has observed about 6 $n/10^6$ α for PuF_4. As a rule of thumb, PuF_4 consisting of 94.5% Pu^{239}, 5% Pu^{240} and 0.5% Pu^{241} yields about 1.5×10^7 n/sec/kg (wt. is of Pu). The increasing α activity with an increasing percentage of Pu^{240} results in greater neutron yields. This is shown in Figure 12. Increased neutron yield because of Am^{241} growth is negligible up to several years after purification. For the above-mentioned mixture, there would be about a 2½% increase at 4 years after purification.

THE GROWTH OF AMERICIUM FROM Pu^{241}

For different applications it may be convenient to express the growth of Am^{241} from Pu^{241} in different ways. Three methods which are commonly used are shown in Figure 13.

TABLE I

NEUTRON YIELDS FROM POLONIUM α PARTICLES ON THICK TARGETS*

Observed Neutron Yield/10^6 α

Target	Roberts (observed)	Roberts (corrected)	Breen and Hertz. (actual sources)
Li_2CO_3	0.66		
Li		2.6	1.0
Be	72	80	59
B	21	24	15
C	0.09	0.11	
N_2	0.01	0.01	
O_2	0.061	0.07	
CF_2	7.5		
CaF_2	5.0		2.8
F_2		12	
Na	1.3	1.5	1.1
Mg	1.2	1.4	0.80
Al	0.64	0.74	0.53
Si	0.15	0.16	
HgCl	0.04		
Cl_2		0.11	
A	0.30	0.38	

Marion, J. B., and Fowler, J. L., Editors, *Fast Neutron Physics*, Chapter 1 A, Interscience Publishers, Inc., New York, 1960.

as curies of Am/g of Pu^{241} with respect to time; grams of Am^{241}/g of Pu^{241} with respect to time; and ppm of Am^{241} in a Pu mixture with respect to time. In the last case, 1% Pu^{241} is assumed to be in the mixture. The plot also assumes that at 0 time, the mixture is void of Am^{241}. For a quick calculation the product 1.44t where t is in days and < 2

years yields the ppm growth of Am^{241} for a mixture containing 1% Pu^{241}. Values for other initial percentages of Pu^{241} would be in direct proportion to the values on the graph.

Of interest also is the decreasing percentage of Pu^{241} with time. Assuming unity at time 0, Figure 14 is a plot of the portion of Pu^{241} remaining as time increases.

FISSION PRODUCT IMPURITIES ASSOCIATED WITH Pu OPERATIONS

Traces of Ru^{103}-Rh^{103} are sometimes found as impurities in Pu. The effective half-life for these isotopes is about 40 days. Gamma energies average about 500 kev. Found more commonly are the impurities Zr^{95}-Nb^{95}. Both Zr^{95} and Nb^{95} contribute gammas of average energy 750 kev. Zr^{95} decays into Nb^{95} with a 65-day half-life and Nb^{95} in turn decays to stable Mo^{95} with a 35-day half-life. The combined activity changing with time, therefore, cannot be represented by a simple exponential. Figure 15 shows this relationship assuming the original activity of Nb^{95} is 0. The equation of this plot is

$$A = e^{-\frac{0.693t}{65}} + \frac{7}{6}(e^{-\frac{0.693t}{65}} - e^{-\frac{0.693t}{35}})$$

with t in days.

SHIELDING DATA FOR MATERIALS COMMON TO Pu AND Am^{241} OPERATIONS

Table II lists the percent transmission through materials common to glovebox construction for the various γ energies usually encountered.
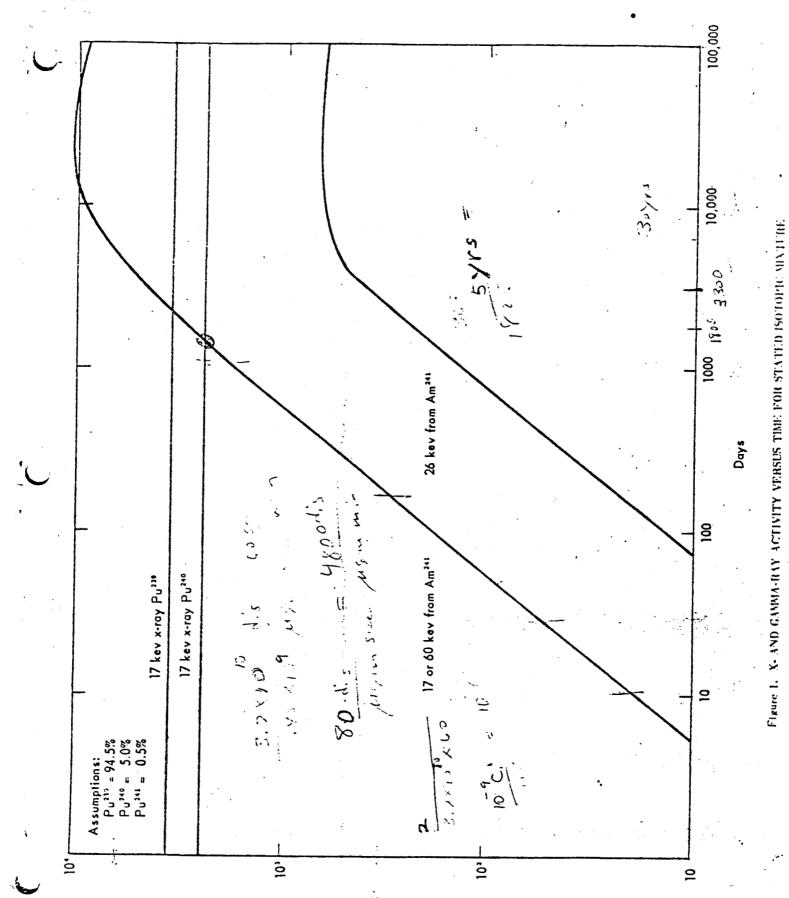
TABLE II

PERCENT TRANSMISSION OF GAMMAS THROUGH SEVERAL MATERIALS

Energy Mev	Pb Gloves* 30-Ga	50-Ga	Pb Glass 4.8 Sp. Gr.** 1/4"	1/2"	3/4"	1"	Safety Glass** 1/4"	1/2"	3/4"	1"	Plexiglas** 1/2"	1"	Lead** 1/16"	1/8"	1/4"	Steel** 1/8"	1/4"	1/2"
0.017	1	0	1.3	0.0			13				40	15	0.0			0		
0.060	85	45	1.5	0.0			75	55	42	32	81	68	0.0			0		
0.100	78	29	5.4	0.0			84	70	58	49	82	71	35.0	10.0	1.0	45	20	3.:
0.200	96	82	33	14.0	5.4	2.2	88	76	67	60	87	76	47.0	25.0	5.0	82	67	45.0
0.380	99	96	69	47.0	31.0	21.0	90	84	75	68	90	86	70.0	56.0	30.0	82	68	47.0
0.510	-100	97	75	60.0	45	35							80.0	70.0	50.0			
0.750	-100	98	81	66	52	45	94	87	80	75	90	86	85.0	78.0	62.0	85	74	56.
1.27	-100	-100	87	74	61	52							88	85	72	90	79	61.
2.1	-100	-100	90	80	73	64							92	87	76	90	80	64.

* 30-Gage gloves ~0.04 mm Pb equivalent
  50-Gage gloves ~0.20 mm Pb equivalent
** Moulthrop, H. A., *Data on Gamma Shielding of Special Plutonium Samples, Z Plant Radiation Study*, Interim Report No. 5 — Part II, HW-61755, Part 2, October 22, 1959.

Assumptions:
$Pu^{239} = 94.5\%$
$Pu^{240} = 5.0\%$
$Pu^{241} = 0.5\%$

17 kev x-ray Pu^{239}

17 kev x-ray Pu^{240}

26 kev from Am^{241}

17 or 60 kev from Am^{241}

x- or γ rays/min/μg of mixture

Days

Figure 1. X- AND GAMMA-RAY ACTIVITY VERSUS TIME FOR STATED ISOTOPIC MIXTURE.

Assumptions:
$Pu^{239} = 94.5\%$
$Pu^{240} = 5.0\%$
$Pu^{241} = 0.5\%$

Pu^{239}

Pu^{240}

Am^{241}

Days

α dpm/μg of mixture

10^6

10^5

10^3

10^1

10^0

10

100

1000

10,000

100,000
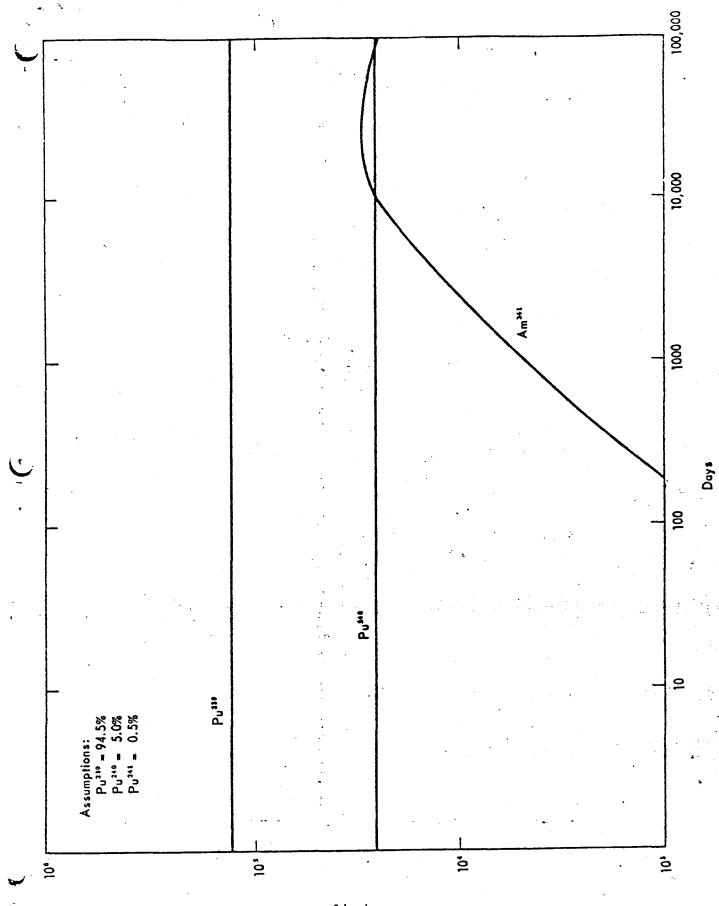
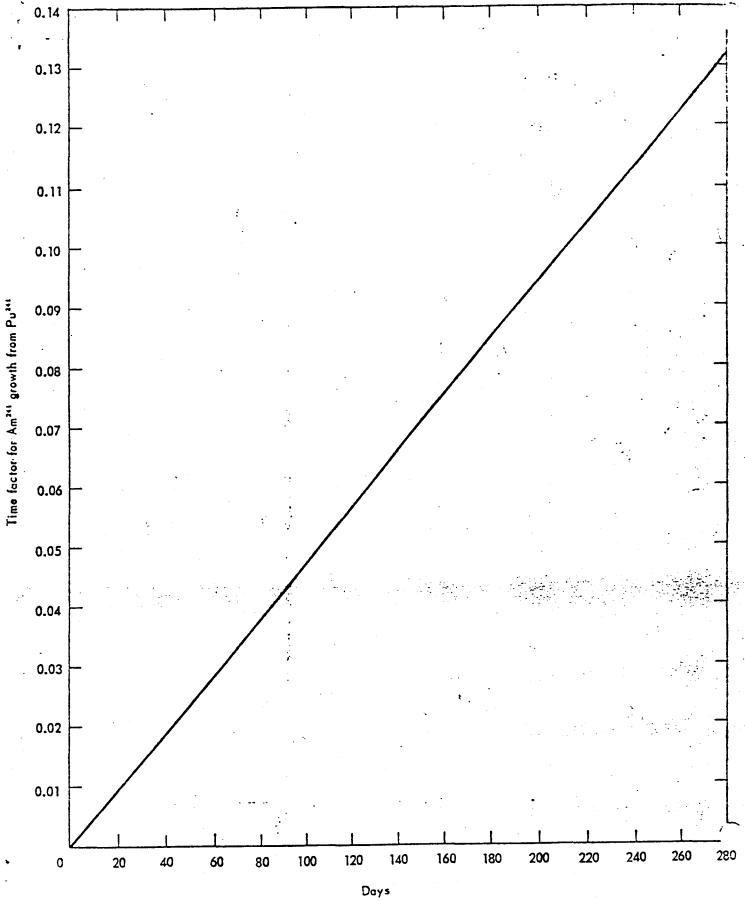Figure 3. ALPHA ACTIVITY VERSUS TIME FOR STATED ISOTOPIC MIXTURE
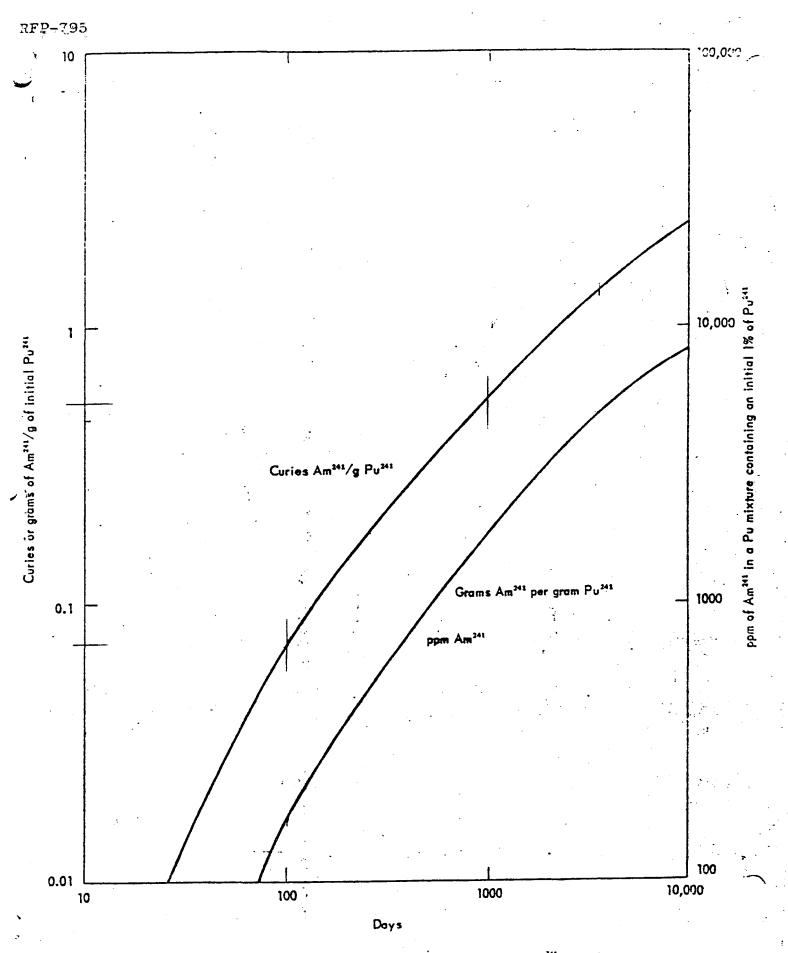
Figure 7. Am²⁴¹ GROWTH FACTOR (use with Figure 8)

Figure 13. Am²⁴¹ CONTENT IN MIXTURE CONTAINING ONE PERCENT Pu²⁴¹ VERSUS TIME